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Experimental Protocols. Volume 1
A Review and Evaluation of Performance

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Volume 1: A Review and Evaluation of Performance

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AIR POLLUTION EXPOSURE SYSTEMS AND EXPERIMENTAL PROTOCOLS:
VOLUME 1: A REVIEW AND EVALUATION OF PERFORMANCE

William E. Hogsett
US EPA Environmental Research Laboratory
200 SW 35th Street
Corvallis, OR 97333

David Olszyk
Statewide Air Pollution Research Center
University of California
Riverside, CA 92521

Douglas P. Ormrod
Department of Horticultural Sciences
University of Guelph
Guelph, Ontario N1G 2W1
Canada

George E. Taylor, Jr.
Environmental Sciences
Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, TN 37830

David T. Tingey
US EPA Environmental Research Laboratory
200 SW 35th Street
Corvallis, OR 97333

ENVIRONMENTAL RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
CORVALLIS, OREGON 97333

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The workshop was successful primarily because of the cooperation and the interaction among the invited participants. We would like to express our thanks to all these individuals for their participation and subsequent review of this document, and especially acknowledge the contributions and helpful guidance of the working group discussion leaders: Dr. Walter W. Heck, Dr. Michael H. Unsworth, and Dr. William E. Winner.

PREFACE

The Forest Response Research Program within the National Acid Precipitation Assessment Program (NAPAP) has requested information and recommendations for standardization of wet and dry deposition exposure facilities, technology, and experimental exposure protocols to investigate the role of atmospheric deposition in forest decline. Standardization of exposure facilities and protocols will permit close comparability among exposure studies conducted on forest species at a variety of sites across the United States. The task of obtaining a collective judgment on the recommendations was best accomplished through an international workshop in which representatives from a large number of laboratories active in pollutant deposition exposure studies participated.

The international workshop was hosted by the U.S. Environmental Protection Agency Corvallis Environmental Research Laboratory on January 27-28, 1986, in Corvallis, Oregon. Prior to the workshop, the authors prepared a review and evaluation of all available wet and dry deposition exposure systems and experimental exposure protocols; the draft document was provided to each participant to serve as a working paper and point of departure for discussion. This review and evaluation, following critical review by the participants and revision, is included in this document as Chapters 1-5 of Volume I and the appendices of Volume II. The consensus view of the participants on the recommended level of standardization of exposure studies with forest species is presented in Chapter 6.

The twenty-five invited participants were asked to work in small, interactive groups to examine and discuss recommendations for standardization within the three main categories: (1) dry deposition exposure systems; (2) wet deposition exposure systems; and (3) deposition exposure regimes. Participants were asked to discuss the following issues: (1) What is the minimum level of standardization of exposure studies (wet and dry deposition) to pursue? (2) Should that level of standardization be applied to specific equipment/facility or should the level of standardization be applied to performance characteristics? (3) What is the recommended standard of exposure system and protocol for specific research objectives? A consensus view on recommendations for each category was established in the larger discussion group, and these are given in Chapter 6. The conclusions of this workshop were initially presented to the NCASI workshop on Controlled Exposure Techniques and the Evaluation of Tree Responses to Airborne Chemicals, held in Atlanta, Georgia, February 4-5, 1986.

International Workshop on Standardization of Exposure Systems and Protocol

Corvallis, Oregon

January 27-28, 1986

PARTICIPANTS

Name/Organization

Wayne L. Banwart
Department of Agronomy
University of Illinois
Turner Hall
Urbana, Illinois 61801

Jeff Brandt
Acid Deposition Staff (RD-676)
U.S. Environmental Protection Agency
401 M Street, S.W.
Washington, D.C. 20460

Philip Dougherty
School of Forest Resources
University of Georgia
Athens, Georgia 30602

Walter W. Heck
USDA/NCSU
Raleigh, North Carolina 27650

W. E. Hogsett
EPA/ERL
Corvallis, Oregon 97333

Patricia Irving
Argonne National Laboratory
Building 203
Argonne, Illinois 60439

Keith F. Jensen
U.S. Department of Agriculture
Forest Service
359 Main Road
Delaware, Ohio

Garret Kats
Statewide Air Pollution
Research Center
University of California
Riverside, California 92521

Lance W. Kress
Argonne National Laboratory
RER/203
Argonne, Illinois 60439

John Laurence
Boyce Thompson Institute
Tower Road
Ithaca, New York 14853

Allen S. Lefohn
ASL & Associates
111 N. Last Chance Gulch
Helena, Montana 59601

Keith Lewin
Department of Energy and Environment
Brookhaven National Laboratory
2 Center Street
Upton, New York 11973

Richard Mandl
Boyce Thompson Institute
Tower Road
Ithaca, NY 14853

Grady E. Neely
EPA/ERL
Corvallis, Oregon 97333

Douglas P. Ormrod
Department of Horticultural Sciences
University of Guelph
Guelph, Ontario N1G 2W1
Canada

Benjamin Stout
NCASI
260 Madison Avenue
New York, New York 10016

Jack Winjum
Weyerhaeuser Company
505 N. Pearl Drive
Centralia, Washington 98531

George E. Taylor, Jr.
Environmental Sciences
Oak Ridge National Laboratory
P.O. Box X
Oak Ridge, Tennessee 37830

David T. Tingey
EPA/ERL
Corvallis, Oregon 97333

Bill Winner
Director, Laboratory for Air Pollution
Impact to Agriculture and Forestry
Department of Plant Pathology and Physiology
Virginia Polytechnic Institute
and State University
Blacksburg, Virginia 24060

George Krause
Landesanstalt für Immissionsschutz
des Landes Nordrhein-Westfalen
Wallneyer Str. 6
4300 Essen 1
West Germany

Lena Skarby
Swedish Environment Research
Institute
P.O. Box 5207
S-402 24 Göteborg, Sweden

M. H. Unsworth
Institute of Terrestrial
Ecology
Bush Estate, Penicuik,
Midlothian, EH26 0QB, Scotland

Mike Ashmore
Department of Pure and Applied Biology
Imperial College at Silwood Park
Ascot, Berkshire SL5 7PY
Great Britain

Hans-Dieter Payer
GSF, Abt. Toxikologie
Ingolstädter Landstrassen 1
8042 Neuherberg
West Germany

I. Introduction

A first step in the process of determining if, and to what level, standardization of exposure systems and protocols should be recommended to the Forest Response Research Program is the integration of current knowledge of exposure facilities, experimental protocols for exposure, and characterization of ambient air quality as it relates to exposure protocol. This document provides both descriptions and evaluations of existing dry and wet deposition exposure facilities, monitoring equipment, and microenvironmental sampling.

The definitions of dry and wet deposition of Fowler (1980) are used in this review. Dry deposition is the direct transfer to and absorption of gases (SO_2 , O_3 , NO_x) and particles by natural surfaces (e.g., vegetation). Wet deposition is the transfer of an element or substance from the atmosphere to the earth's surface within, or on the surface of, a hydrometeor (e.g., rain, snow, fog, mist, aerosol).

Information on the various exposure systems has been organized based on the degree of environmental control the various systems are capable of, rather than the degree of pollutant control the systems can accomplish (e.g., controlled-environment exposure chambers, greenhouse exposure chambers, open-top chambers). The types of systems are discussed and evaluated as a group; detailed descriptions of each facility are given in the appendices. The equipment, including the various types of chambers for both wet and dry deposition, pollutant monitoring, pollutant-dispensing and control, and microenvironmental data acquisition are evaluated using generic criteria. Experimental protocols for exposure dynamics are discussed and a review of ambient air quality and its characterization as related to wet and dry deposition exposure studies is included. Recommendations are given for exposure regimes for vegetation effects studies.

A descriptive evaluation of the exposure system performance is outlined using generic criteria which reflect conditions necessary to provide realistic and reproducible experimental conditions. The criteria are related to the pollutant chemistry in the atmosphere and the plant's edaphic, climatic, and atmospheric environment during both exposure and non-exposure periods. The evaluation is descriptive, and includes a discussion of each system's strengths and limitations.

The relative importance of the criteria will vary, depending on research task objective. Thus the evaluation of available research systems and final recommendations should be tempered with the understanding that criteria stringency depends on the biological objective that the facilities or protocols were originally designed to meet. In addition, the selection of appropriate criteria for evaluating the various systems, the evaluation of the systems, and finally, recommendations regarding standardization are also tempered with consideration of these exposure systems for use with forest tree species since most were developed for use with crop species and have followed protocols directed towards annual and perennial crop species. In discussion and consideration of exposure systems and protocol, the specific requirements for tree species are of primary importance in arriving at final recommendations of exposure systems and regimes.

Experimental tree studies present the obvious problem of size, of both height and canopy coverage; and, also, increased variability; slower growth rates compared to annual crops; length of exposure period required; and in the case of certain conifers distinct growth "flushing" periods. Additional, but no less important considerations, have been outlined by Cowling (1985, personal communication):

1. "Forest trees are perennial plants which accumulate biomass 12 months a year, even in northern latitudes. Roots of forest trees grow whenever the ground is not frozen. A very important part of net photosynthesis in many forest trees occurs during the "dormant season," even in deciduous forests."
2. "After the first year of growth, most coniferous trees carry from as few as two to as many as ten or even twelve different year-ages of needles which differ in physiological activity and susceptibility to stress. Thus, responses to stress in young seedlings with only primary needles or only one-year-old and/or two-year old secondary needles may not be the same as responses of seedlings, saplings, or mature trees with many different year-ages of needles."
3. "Loss of foliage due to premature loss of needles may be of no importance in determining net photosynthesis in forest stands because of the redundancies and normal shading effects within forest canopies."
4. "Roots of forest trees extend far beyond (often 3 to 4 times) the diameter of the aboveground canopy."
5. "Forest trees normally allocate a significant part of current photosynthesis to synthesis of new feeder roots and to maintaining symbiotic associations with mycorrhizal fungi. These so-called "root turnover" or "root maintenance," and/or "mycorrhizal maintenance" functions constitute a large drain (10-40% of gross photosynthesis) on the energy resources of the trees. Thus, marginal impairment of photosynthesis by airborne chemicals or other stress factors can result in nutrient deficiencies induced by roots that are not fed adequately and therefore do not perform their nutrient-uptake responsibilities adequately."
6. "Trees grow in diameter long after they stop growing in height."
7. "Some forest trees have multiple flushes of shoot growth during the same growing season which are determined by different sets of buds."
8. "The capacity of some forest trees to grow in height during a given growing season is determined by the nature and vigor of buds set during the previous growing season."
9. "Symbiotic associations between forest trees and soil microorganisms are exceedingly important determinants of growth."

10. "Forest soils are essentially never limed or irrigated and rarely cultivated or fertilized. Thus, forest trees usually experience some nutrient and water stress during most growing seasons."
11. "Forest soils are normally very heterogeneous."
12. "Most forests (even in the southern pine region) are wild ecosystems which do not depend on man for their perpetuation."
13. "The natural range (and therefore the ecotypic and genetic diversity within) many forest tree species is very much larger than the natural range, ecotypic variability, and genetic diversity of many agricultural crops. Wheat and loblolly pine are good examples. Although wheat is grown all over the world, its natural range was confined to a region of the Near East which is very much smaller than the natural range of loblolly pine."
14. "Forest trees are never free of competition from lesser vegetation."
15. "The net economic values of forest trees at time of harvest could be increased by air pollution if the principal effect were to increase the mortality of suppressed trees in forest stands."

Finally, recommendations are offered by the workshop participants on the minimum level of standardization for exposure facility types, monitoring hardware, and the exposure protocol. The recommendations reflect the issues discussed with respect to system evaluations, the evaluation criteria, and the special concerns of the use of these systems and protocols with forest tree species.

II. System Descriptions

All atmospheric pollution-vegetation studies are designed to address specific research objectives at the biochemical, physiological, whole plant, or population level. The facility used depends upon the objective; systems are designed to control the pollution environment to test hypotheses and to minimize the facility effect on the environment during the pollution event. Although the systems discussed in this document were originally designed primarily to address whole plant and population effects, all have been used in experiments designed to address various biochemical and physiological hypotheses.

A. Dry Deposition Systems

Most facilities for investigating the effects of dry deposition on vegetation have been designed to study gaseous air pollutants. Very few facilities have been designed to evaluate the effects of particle deposition, in terms of either ambient exposures or controlled addition.

1. Gaseous Exposures

Facilities available that accommodate experimentally controlled exposures with gaseous pollutants can be divided into two main types based on degree of manipulation of the atmosphere and plant environment: (1) non-chamber systems which provide a degree of pollutant exposure control with no appreciable alteration of the plant environment; and (2) chamber systems which provide much greater control of the pollutant exposure but modify the environment somewhat from ambient conditions. A third method, infrequently used, is the ambient gradient. It incorporates known differences in pollutant doses in the field, but does not allow for manipulation of ambient to create a range of treatments (S. V. Krupa, personal communication; Colvill et al., 1985; Oshima et al., 1976). The ambient gradient procedure is not discussed in this report.

a. Non-Chamber Systems

Non-chamber field exposure facilities are of two main types: (1) systems designed to emit a plume of pollutant over a plant canopy and thus dependent on ambient wind for diffusion and dispersion; and (2) systems designed to exclude ambient air from plants by blowing filtered air over a canopy during fumigation episodes, possibly coupled with pollutant addition.

(1) Plume Exposures

All plume systems are designed to treat target plants with pollutants added above background levels. The pollutant is injected into ambient air and relies on local winds for dispersion. Very simple emitters, e.g., thin tubes, are used, resulting in no facility effect on the plant environment.

The first plume systems used an array of tubes throughout a plot to provide exposure regardless of wind direction. One system was designed to expose native grasses to SO₂ fumigations similar to those possible from an electric generating point source (Lee et al., 1975). This Zonal Air Pollution

System (ZAPS) consisted of a network of long and short aluminum pipes suspended 0.75 m above 0.5-ha plots. Gas release holes were positioned at 3-m intervals with more than 250 points above the canopy. The design consisted of a series of plots, each receiving a long-term exposure to a different concentration of SO₂. The rate of SO₂ release was constant, resulting in high variability in horizontal and temporal concentrations with different wind speeds and directions.

Another early plume system used an array of 128 vertical tubes distributed through an exposure plot, with each tube emitting SO₂ at various heights (de Cormis et al., 1975). This system provided for low variability in vertical pollutant concentrations for the tree species studied. However, horizontal variability across the plot and temporal variability were substantial.

Further refinement of plume systems emphasized alteration of the original designs, either to increase the uniformity of pollutant concentrations across the exposure plots and/or to use the variability across plots for a gradient of pollutant exposure concentrations for dose response studies. To alleviate problems and increase potential uses for plume systems, researchers have simplified the array of tubes (Thompson et al., 1984; Miller et al., 1980; Greenwood et al., 1982), devised more efficient shapes for the exposure systems to consider variable wind directions, i.e., circular (McLeod et al., 1985) or square (Greenwood et al., 1982), increased the distance between tubes and target plants (McLeod et al., 1985), or increased system portability (Northrop, 1983). Recent designs have added computer-controlled pollutant dispensing and monitoring components to regulate exposures based on wind speed and direction and to improve the uniformity in concentrations across plots. Several plume systems are described in greater detail in Appendix A.

(2) Air Exclusion

The second type of non-chamber exposure system is the air exclusion facility which permits clean air treatments or removal of ambient air pollutants without the environmental modifications associated with chambers. Most of these systems use inflatable PVC ducts lying between plant rows to blow filtered air over the plants during ambient fumigation episodes. The duct air prevents the incursion of ambient air into the plant canopy and delivers air filtered by charcoal to the canopy. Air exclusion systems are intended to impose negligible alteration of the plant environment since the ducts are inflated and lie alongside the plants only during fumigation. When fumigation is not occurring, the ducts lie flat on the ground, resulting in little or no effect on light, air temperature, humidity, and other climatic variables.

The first air exclusion system was designed by Jones et al. (1977) to exclude SO₂ from soybeans during fumigation episodes from a coal-fired electrical generating station. The system has ducts with holes positioned to release filtered air over a soybean canopy whenever a dedicated SO₂ monitor indicates a fumigation event. The system excludes most of the SO₂ from the top, but is less successful in excluding SO₂ from within the canopy.

Further development of the air exclusion system involved alteration of the number and size of holes, number of ducts, air flow rates, and other parameters to provide for pollutant addition as well as exclusion. Shinn *et al.* (1977) developed a modified system for adding O_3 and H_2S to a plant canopy. The system not only excludes ambient polluted air, but also provides for exposure to a linear gradient of O_3 and H_2S concentrations along the length of the ducts. The gradient is achieved by injecting the air at the blower end of the system and allowing the gradually decreasing emission rate along the length of the ducts to lower the adjacent plot concentration; and by increasing the hole diameter along the length. The exposure plot is surrounded by a 0.6 m high fiberglass barrier to minimize outside air incursion across the plot. This use of the air exclusion system for a linear gradient exposure necessitates extensive air monitoring across the plot to determine plant exposure concentrations for dose-response regression analysis (Bennett *et al.*, 1980; Coyne and Bingham, 1978).

Further development has emphasized changes in design to provide for a variety of pollutant interaction exposures. Laurence *et al.* (1982) refined the system to provide for gradients of HF, and SO_2 plus O_3 . Thompson and Olszyk (1985) designed a system adaptable for a variety of uses including ambient air exclusion, or linear gradients of ambient air or SO_2 . Kuja *et al.* (1985) modified their system to exclude ambient ozone and added a rain cover to prevent ambient rainfall. Air exclusion systems at particular locations are described more fully in Appendix B.

b. Chamber Systems

A wide variety of chambers have been used to provide controlled exposures to defined pollutant doses. The most generalized distinction is (1) outdoor chamber and (2) indoor chamber. Outdoor chambers are designed to control the chemical or pollutant atmosphere around the plants with minimal modification but no direct control of the environment. Indoor chambers control both the chemical or pollutant atmosphere, and the environment, thus minimizing all possible nontreatment effects on plant responses.

The general requirements for all types of chambers are similar (Guderian, 1985; Heagle and Philbeck, 1978). Both outdoor and indoor chambers are designed to: (1) provide environmental and exposure conditions representative of outdoors; (2) provide an environment as uniform as possible within and between chambers; (3) provide for uniform concentrations of defined pollutants both within and between chambers; (4) have transparent coverings to minimize reductions in irradiance quantity and quality; (5) contain exposed surfaces made of nonreactive materials; (6) facilitate experimental manipulation of plants; and (7) be easy to build and maintain. No single design has attained the ideal for all these considerations; each has particular characteristics based on the research goals, available technology and materials, and resources of individual investigators.

(1) Outdoor Chambers

The outdoor or field chamber was originally designed to exclude ambient air while blowing filtered air over the plant canopy. The earliest chambers in use from the early 1950's to early 1970's were closed systems that were essentially small greenhouses (e.g., Thompson and Taylor, 1966; Van Haut, 1972). Some of the chambers were large enough to be used over young citrus trees (Thompson and Taylor, 1969); others were smaller, appropriate for plants ranging from tree seedlings (Miller and Yoshiyama, 1973) to sweet corn (Heagle et al., 1972). All of these early closed chambers were designed to dispense air through the chamber; however, environmental conditions in the chamber did not represent ambient. Increased air temperatures were especially apparent. Air flow through the usually cube-shaped chambers was not necessarily uniform, creating dead air spaces generally in corners.

Current designs of outdoor chambers range from minimal low chambers with open tops to large fully enclosed domes. The majority incorporate features to meet requirements (a) through (f) above, but with features important for a certain type of crop. A versatile 2.4 m high, 3.0 m diameter, PVC-covered open-top chamber designed by Heagle et al. (1973) has been adopted for studies investigating the effects of both ambient and added air pollutants on a wide variety of crops as part of the U.S. Environmental Protection Agency National Crop Loss Assessment Network (NCLAN) (Heck et al., 1982; 1984). Either filtered or ambient air can be blown into the chamber through a perforated duct (plenum) around the inside of the base of the chamber. Air moves up through the plant canopy and out the chamber top where it restricts the incursion of ambient air. A frustrum or baffle around the top of the chamber further restricts air incursion. The open top allows ambient rainfall, minimizes ambient light restriction, helps prevent air temperature increases, and permits entrance of ambient pests.

The essential components of all open-top chambers include: (1) chamber covering permitting light penetration; (2) a high capacity blower to inject air; (3) charcoal and/or particulate filters to remove ambient pollutants; and (4) some structure for dispersing and mixing air entering the chamber, e.g., a plenum. In general, the materials must be durable and resistant to pollutant-induced breakdown. The filters must have a high filtering efficiency for the pollutants in question and, in addition, the filters may have to be chemically treated to remove gases such as SO_2 or NO_x .

Open-top chambers have been used most extensively with herbaceous crops during the spring-summer growing season. The environmental modifications that do occur may limit their usefulness in areas with environmental extremes and during other seasons, especially winter. However, there have been few year-round growth studies in chambers and non-chamber field plots to evaluate the effect of chamber environmental modification. The size and dimensions of the originally-designed chambers have also limited their usefulness for some crops, especially larger trees. In addition, the limited incursion of ambient air that occurs even with the highest air flow rates can affect their usefulness in more polluted areas or if a very clean treatment is required. Low height chambers and/or high air flow rates can reduce the chamber effect on the environment for certain low-growing winter crops such as barley and ryegrass

(Fowler, personal communication; Roberts et al., 1983). Larger chambers can allow space for growth of small trees or large vines (Kats et al., 1985; Heck et al., 1985; Brewer, 1983). Other variations of the chamber design are possible. Outdoor chamber systems for gaseous dry deposition research including open-top, semi-open-top, and closed-top designs are described in Appendix C. Outdoor chambers for studies of gaseous dry deposition and wet deposition combinations are described in Appendix D.

(2) Chambers -- Indoors

The other general chamber type, indoor chambers, has been widely used in air pollution research. These chambers may be used in greenhouses in which natural light is the only or principal light source or in growth rooms in which all the light is artificial. Greenhouse chambers may be units within a greenhouse or entire small greenhouses. The growth room chambers may be units within a growth room or entire rooms. The chambers differ in size, number of integrated units, air circulation patterns, air pressure, air mixing, air exchange rates, and other features. Descriptions of many indoor chambers for gaseous dry deposition research are given in Appendix E.

Early greenhouse chambers used entire greenhouses or exposure chambers (Berry, 1970; Hill et al., 1959). A later version featured several small greenhouse-like structures within a large greenhouse (Posthumus, 1978). Other scientists placed several transparent chambers within a greenhouse (Hill et al., 1959; Lockyer et al., 1976; Piersol and Hanan, 1975). Some designs of exposure chambers have been used to construct multiple units in greenhouses and in growth rooms (Heck et al., 1968; Heck et al., 1978).

Early growth room chambers were converted walk-in storage rooms with artificial lights added (Menser and Heggestad, 1964). Other researchers divided walk-in rooms to permit direct comparisons of control and treated plants (Adams, 1961). Some investigators converted commercial growth chambers into exposure chambers by adding corrosion-resistant coatings and sealing (Wood et al., 1973). Another approach has been to place exposure chambers within commercial growth chambers (Cantwell, 1968; Heck et al., 1968) or within walk-in rooms (Heck et al., 1968; 1978; McLaughlin et al., 1976; Payer et al., 1985). Other investigators have constructed self-contained growth chamber-like units specifically designed for air pollution studies (Hill, 1967; Oliva and Steubing, 1976). In recent years, some very sophisticated indoor ("air pollution phytotron") systems have been constructed (Aiga et al., 1984; Payer et al., 1985) which provide wide ranges of environment and pollutant concentrations that are electronically monitored and controlled.

Many chambers used in air pollution research were designed to be used in groups for experiments. Each group typically has a single air conditioning system and pollutant dispensing and monitoring system. The air exchange is usually based on a single pass-through of conditioned air to which pollutant is added in particular chambers as dictated by the experimental design. There is no extra mixing within the exposure chamber; the speed of air movement through the chamber is assumed to be sufficient to maintain adequate levels of carbon

dioxide and pollutant in the vicinity of the plants. The chambers designed and constructed by Heck et al. (1968), Jensen and Bender (1977), and Lockyer et al. (1976) are of this type.

The continuous-stirred tank reactor (CSTR) exposure chamber was developed to permit plant growth and gaseous pollutant studies using the principles of chemical reaction systems (Rogers et al., 1977). A set of two chambers, mounted on a cart, was constructed and tested in a walk-in plant growth room in the North Carolina State University Phytotron. The thorough mixing in the CSTR chamber ensures that plants are exposed to the concentration of pollutant measured at the outlet and that leaf boundary layer resistance is small. Measurement of inlet and outlet pollutant, carbon dioxide, and water vapor concentrations permit calculations of pollutant flux, net photosynthesis, and transpiration rates, respectively. This concept for exposure chambers was extended to the design and construction of a nine-unit greenhouse and four-unit growth room (Phytotron) facility (Heck et al., 1978). Subsequently, CSTR chambers have been constructed at several locations. Many of the CSTR systems have been developed to control temperature and humidity as well as provide high light levels.

There are many other types of controlled environment indoor facilities in use. Some are quite sophisticated but have not been the subject of methods publications. Such chamber systems are sometimes described in papers dealing largely with research results. The chamber systems described in this section thus represent only examples of the array of chamber systems in use today.

(3) Cuvettes

Use of leaf cuvettes in air pollution research has been limited, even though they are widely used by plant physiologists in leaf photosynthesis studies. Photosynthesis research can help clarify plant-environment relations, is fundamental in the analysis of carbon metabolism, and can help define physiological mechanisms which alter plant growth rates and form (Mooney, 1972a). A basic introduction to principles of gas exchange studies is presented by Sestak et al. (1971). Cuvettes in current use are generally designed to monitor gas fluxes on a leaf area basis for an individual leaf, or leaf part, while it is still attached to the plant. Although whole plant and multiple plant chambers can be used for gas exchange studies, these facilities are described elsewhere. Cuvettes must have gas handling systems and monitors for water vapor, CO₂, and possibly air pollutants. Integration of these components constitutes a gas exchange system.

Cuvette systems used in air pollution experiments are commonly operated independently from exposures, i.e., gas exchange rates are determined before or after fumigations, but fumigations do not occur within the cuvette. Cuvettes in this case can be used to document the effects of gaseous air pollution exposures (e.g., McLaughlin et al., 1979; Norby and Kozlowski, 1981; Olszyk and Tibbitts, 1981; and Kimmerer and Kozlowski, 1981).

A few systems exist in which air pollutants can be introduced directly into the cuvette. These systems constitute air pollution exposure systems and can be used to calculate air pollution uptake characteristics of single leaves

as well as dynamic physiological responses of leave to exposure. The most common design is the single pass, flow-through system which operates by injecting air pollutants into the air stream before gases flowing through the system enter the cuvette (Black and Unsworth, 1979; Taylor and Tingey, 1979; Winner and Mooney, 1980; Hallgren et al., 1982; Noble and Jenson, 1983; Atkinson, 1986). Thus, monitoring air pollution concentrations in the air streams entering and leaving the cuvette allows the device to be used as an air pollution exposure chamber. Studies with SO_2 have led to approaches for calculating total SO_2 uptake by leaves and for partitioning total uptake between the fraction adsorbed to leaf surfaces and the fraction adsorbed into leaf mesophyll (Taylor and Tingey, 1979; Winner and Mooney, 1980b,c). These approaches used for analysis of SO_2 uptake should be appropriate for the analysis of fluxes of other air pollutants, such as O_3 , between the leaf and air.

There are four basic approaches to gas exchange studies in cuvettes. All of these cuvette systems are commercially available; however, many scientists develop their own equipment or modify commercial units.

1. Cuvettes for Sampling Rates of Gas Absorption or Emission from Leaves. These cuvettes are generally small chambers with a known area volume and they readily clip on to a portion of a leaf of known area. There is no air flow through the cuvette but a fan is used to mix air. If the cuvette contains a humidity sensor and thermocouples to measure leaf and air temperatures, the rate of increase in relative humidity in the cuvette is equated to the leaf transpiration rate and this value can be used to calculate stomatal conductance (Sestak et al., 1971). Newer systems simultaneously monitor rates of increasing water vapor concentration and decreasing CO_2 concentration in the cuvette which allows calculation of transpiration, stomatal conductance, and photosynthesis. These systems are relatively inexpensive, easy to use, and field portable (Winner and Mooney, 1980a). The cuvettes are simple and lack environmental control. Gas exchange rates are measured quickly (less than 2 minutes) to prevent leaf heating, the extreme buildup of water vapor, or CO_2 depletion. Experiments generally involve determining gas exchange rates of many leaves (reference?). These cuvettes can be adapted to collected gases emitted from leaves, e.g., ethylene (Tingey et al., 1976) and H_2S (Winner et al., 1982).

2. Cuvettes for Continuous Measurement of Gas Absorption or Emission from Leaves -- Differential Gas Exchange Systems. These cuvettes commonly are used to study gas exchange of an entire leaf while still attached to a plant. The cuvette is equipped with devices to control temperature, photosynthetically active radiation, and air flow rates, offering a high degree of environmental control (Black and Unsworth, 1979; Legge et al., 1977; Linder et al., 1980; Winner and Mooney, 1980b). This is necessary because of the long duration (1-8 h) of experiments with single leaves. Experimental air is preconditioned to desired temperature and water vapor, CO_2 , and perhaps air pollutant content. The air stream is split into reference and sample lines. The reference line is directed to sensors for water vapor and CO_2 analysis and the sample line is directed first to the cuvette and then to gas analysis sensors. The cuvette operates on a flow-through basis, i.e., gases pass through the cuvette only once. Differences in water vapor and CO_2 concentrations between the sample and reference lines are equated to transpiration and photosynthesis. These data

can be used to calculate a number of important physiological parameters including stomatal conductance and partial pressures of CO_2 in mesophyll tissues (Jarvis et al., 1971) and gaseous air pollution flux into leaves (Unsworth et al., 1976; Winner and Mooney, 1980b,c).

Monitoring and control of all important environmental factors allow study of dynamic leaf responses to gaseous pollutants such as SO_2 under constant conditions so that results from many experiments can be compared (Winner and Mooney, 1980d; Winner et al., 1982). The interaction of gaseous pollutants with environmental factors can also be studied (Norby and Kozlowski, 1981; 1982; Jones and Mansfield, 1982). These systems are often used to determine responses of leaves to wide ranges in light, CO_2 , and vapor pressure deficit. The resultant response curves are useful for diagnosis of quantum yield and intrinsic photosynthetic capacity of mesophyll cells. Differential gas exchange systems are usually developed for laboratory studies and used in conjunction with computerized data acquisition systems. Recent engineering advances now make these systems field portable. They are relatively expensive and require technical expertise (Oechel and Lawrence, 1979; Mahon and Domey, 1979; Lange and Tenhunen, 1984; 1985).

3. Cuvettes for Maintaining Constant Levels of Water Vapor and CO_2 -- Null Balance, Steady State Gas Exchange Systems. Null balance systems are commonly used to survey gas exchange rates for many leaves and in this application, temperature and radiation are monitored but control of these environmental factors is unnecessary (Nilson et al., 1984). This approach to gas exchange involves monitoring water vapor and CO_2 in the air around a leaf contained within a flow-through cuvette. Dry air or CO_2 are added to the air stream to bring ambient humidity and CO_2 to desired levels. Transpiration rate is calculated from the rate at which dry air is added to the cuvette to reduce humidity to a predetermined level. Photosynthesis rate is calculated from the rate at which CO_2 is added to increase CO_2 concentrations in the cuvette to predetermined values. Predetermined humidity is often the level outside the cuvette and predetermined CO_2 is often 320 ppm. These systems are commercially available, relatively expensive, field portable, and easy to use (Coyne and Bingham, 1981).

4. Cuvettes for Carbon Isotope Studies. Cuvettes are used to expose single leaves to CO_2 containing unstable carbon isotopes. Radioactivity emitted during decay of ^{11}C and ^{14}C can be monitored to document rates of photosynthesis and photosynthate translocation from source leaves to other plant tissues. $^{14}\text{CO}_2$ experiments require pulse-chase techniques and destructive sampling (Austin and Longden, 1967) to detect ^{14}C with scintillation techniques (Smith, 1969). The nature of pulse-chase experimentation and techniques for generating $^{14}\text{CO}_2$ make control of CO_2 concentrations in the cuvette difficult. Most cuvettes used in radiolabeling experiments are simple and have no environmental control. The $^{14}\text{CO}_2$ techniques can be used in the field research (Tieszen et al., 1974; McLaughlin et al., 1982). Although this research approach can illustrate photosynthesis and allocation, it is difficult to detect the dynamics of plant responses to air pollutants. In addition, $^{14}\text{CO}_2$ studies do not describe stomatal conductance, and photosynthesis rates measured with $^{14}\text{CO}_2$ techniques may not match rates measured by infrared gas analysis techniques (Karlsson and Sveinbjornsson, 1981). The $^{11}\text{CO}_2$ experiments

are nondestructive since the ^{11}C can be monitored throughout the plant with geiger counters. The $^{11}\text{CO}_2$ experiments can reveal dynamic changes in photosynthesis and carbon allocation in environmentally controlled studies, but this research is expensive and can only be done at a few locations (Magnuson et al., 1982).

2. Non-Gaseous (Particulate) Exposures

There have been few studies on the effects of dry particulate on plants under controlled exposure conditions. Requirements for quantitative and uniform applications pose special problems in chamber design and experimental methodology. Particle size characterization is required along with quantitative and repeatable application methods.

Several non-gaseous dry deposition studies have been conducted outdoors. For example, Chamberlain and Little (1981) described the nature of the particulate pollutant deposition process by studying radioisotope and lead contamination of plants set outdoors in the zone of influence of the contaminants. Other studies have been conducted in more closely controlled conditions indoors. Plant exposure chambers for dust studies were described by Darley et al. (1968) who were studying cement-kiln dust effects on plants. Krause and Kaiser (1977) dusted their plants with heavy metal particulate for studies of metal- SO_2 interactions. The "special dusting chamber" was not described. Ormrod et al. (1986) devised a chamber for metal dust application to plants based on the chamber designed by Marple and Rubow (1983) for studies of dust effects on electronic instruments. Several chambers for particulate dry deposition research are described in Appendix H.

3. Pollutant Monitoring

The gaseous air pollutant monitoring program will need to follow approved Quality Assurance Guidelines. This will assure provision of air pollutant analyzer data which is of known quality and accuracy and is therefore legally defensible. The National Crop Loss Assessment Network (NCLAN) Quality Assurance Plan for Air Pollutant Analyzers (dated September 11, 1985, U.S. Environmental Protection, Corvallis, Oregon) identifies procedures which are necessary to air pollutant analyzer measurement and calibration activities. All analyzers used in the NCLAN project for measuring pollutant levels in the ambient atmosphere must use a Reference Method and meet the equivalency requirements specified by EPA in the Federal Register, 40 CFR 53.

Monitoring of pollutants involves many concerns and precautions including sampling locations, lengths and characteristics of sampling lines, location of analyzers, and frequency of sampling (Tibbitts, 1978). In most experiments involving several treatment levels, time-sharing of monitoring instruments may be required. The number of locations that can be sampled with a single instrument is dictated by the response time of the monitor and the degree of variability expected in the pollutant concentration over exposure time (Heagle and Philbeck, 1978). Sampling should be frequent enough to ensure use of monitoring data for the various exposure statistics dictated by the research objective. Time-sharing systems usually involve continuous flow sampling lines, a sampling manifold, a scanning valve control unit, and monitoring

instruments for each gas under study. Each monitor draws samples directly from the sampling manifold, allowing simultaneous analysis of different pollutants. The monitoring instruments must be able to analyze continuously and have a fast response time with low sample air flow rate.

Analyzers for ozone, sulfur dioxide, and nitrogen oxides are classified by detection method. Table II-1 compares analyzers for the three gases with respect to various performance parameters and cost (Burmahn and Rehme, 1978). These tables indicate that only some analyzers will be acceptable and useful in experimental work involving the need for rapid and accurate monitoring of pollutant gases.

It should be noted that some of these instruments have been modified and improved since publication of these tables. For example, the response time of pulsed UV fluorescent SO₂ analyzers can be reduced to about 90 sec by modifying standard instruments; the lower detectable limit for this type of analyzer should be lower than 10 ppb. Other considerations for selection of analyzers include size, weight, portability, power source and consumption, and requirement for accuracy gases.

4. Pollutant Dispensing and Control

The basic requirements for dispensing systems for gaseous dry deposition have been described by Heagle and Philbeck (1978) (Appendix I). According to them, an acceptable dispensing system: (1) delivers easily-controllable concentrations of pollutant to one or more exposure chambers; (2) is free of leaks; (3) is relatively free of concentration "drift"; and (4) is constructed with nonreactive materials to prevent corrosion and the production of secondary substances. A system that dispenses pollutants to several chambers at once is usually preferable to separate systems for each chamber. Each dispensing system consists of three major parts: (1) a pollutant source (usually compressed gases or an ozone generator); (2) a series of precision valves or flowmeters; and (3) an air dilution manifold that carries the gas to the inlet ducts. Field dispensing systems require greater air-moving capacity than those for greenhouse or laboratory chambers. Larger initial pollutant concentrations in cylinders or at the generator are usually required to compensate for the high volume of air moving through field systems.

Generalized pollutant control systems have also been described by Heagle and Philbeck (1978). The type of control may depend on the experiment objective: a single static set concentration may be used; several set concentrations varying with time may be used in one or more chambers; or gradually changing dynamic concentrations may be used. A simple control may consist of solenoid valves which are turned on by a microswitch controller mounted on a recorder. A set of concentrations is added and once flow rates and pollutant concentrations are adjusted manually to near the desired levels, the method is satisfactory. A more elaborate system, in which valves open a proportional amount, uses "servo" valves to maintain the concentration more smoothly over time. Programmable control systems are available for dynamic dose-response studies. A monitoring instrument and a programmed exposure regime are compared electronically and the volume of pollutant injected into the chamber is varied accordingly.

Table II-1. Comparisons of analyzers with respect to major performance parameters and procurement cost (from Burmann and Rehme, 1978).

TYPICAL OZONE/OXIDANT ANALYZERS

Parameter	Analyzer				
	Colorimetric	Gas-Phase Chemiluminescent	Gas-Solid Chemiluminescent	UV Photometric	Electrochemical
Lower Detectable Limit	10-20 ppb	< 1-10 ppb	< 1-10 ppb	< 1-10 ppb	10-20 ppb
Specificity ^a	low	high	high	high	low
Stability ^b	low	high	high	high	low
Response Time	< 5 min	< 1 min	< 1 min	< 1 min	< 5 min
Working Range	up to 10 ppm	up to 2 ppm	up to 2 ppm	up to 10 ppm	up to 10 ppm
Cost	3-6 K	3-6 K	> 6 K	3-6 K	1-3 K

TYPICAL SO₂ ANALYZERS

Parameter	Analyzer						
	Conducti- metric	Voltametric	Amperometric	Colorimetric	Flame Photometric	2nd Derivative UV Spectrometric	Pulsed UV Fluorescent
Lower Detect- able Limit	10-20 ppb	10-20 ppb	10-20 ppb	10-20 ppb	< 1-10 ppb	10-20 ppb	< 10 ppb
Specificity ^a	moderate	moderate	moderate	high	moderate	high	moderate
Stability ^b	low	low	high	low	high	high	high
Response Time	< 5 min	< 5 min	< 5 min	< 5 min	< 1 min	< 5 min	< 5 min
Working Range	up to 10 ppm	up to 10 ppm	up to 2 ppm	up to 4 ppm	up to 10 ppm	up to 2 ppm	up to 5 ppm
Cost	1-3 K	1-3 K	3-6 K	3-6 K	3-6 K	> 6 K	> 6 K

TYPICAL NO_x ANALYZERS

Parameter	Analyzer			
	Voltametric	Amperometric	Colorimetric	Chemiluminescent
Lower Detectable Limit	10-20 ppb	10-20 ppb	10-20 ppb	< 1-10 ppb
Specificity ^a	low	low	high	low
Stability ^b	low	low	low	low
Response Time	< 5 min	< 5 min	> 5 min	< 1 min
Working Range	up to 10 ppm	up to 12 ppm	up to 2 ppm	up to 10 ppm
Cost	1-3 K	3-6 K	3-6 K	< 6 K

^a Specificity: high -- < 10% error from species commonly encountered in ambient air; moderate -- scrubber required to eliminate interferences; low -- scrubber may not eliminate interferences under all conditions, and/or data corrections required based on concurrent measurements.

^b Stability: high -- meets EPA Reference and Equivalent Method specifications; moderate -- may be operated without significant drift for 1-2 days; low -- requires daily zero/span adjustment.

The technology available for pollutant dispensing and control is changing rapidly, and new, more efficient, precise systems are becoming available. For example, the use of a combination of a microcomputer and electronic flow controller can provide complex dynamic concentration regimes in both outdoor and indoor research facilities (Hogsett et al., 1985; personal communication).

A varied array of pollutant control systems has been described, usually in conjunction with descriptions of exposure systems. More details and references are included with facility descriptions in the appendices.

5. Environment Monitoring

Detailed and complete measurements of the environment are needed because environment interacts with pollutant response and because of the differences in exposure facility designs, surrounding environmental conditions, and cultural practices in different facilities (Mandl et al., 1973; Heagle and Philbeck, 1978; Tibbitts, 1978; Olszyk et al., 1980; Unsworth et al., 1984a,b). Despite efforts to standardize the environment, existing differences can create significant variations in irradiance patterns, airflow patterns, atmospheric composition, temperature patterns, and other factors which may affect pollutant response. Inadvertent environmental stresses created by subtle factors such as water and nutrient supply, excessive vibration or wind action, and unplanned contaminants also may adversely affect plant response. Thorough environmental measurements and detailed reporting will help identify interactions affecting response variables.

Guidelines providing a common basis for the measurement of environmental parameters promote a uniform, accurate means of reporting data and results from air pollution experiments. Such guidelines have been prepared by a panel of plant scientists and published in several journals including HortScience, a journal published by the American Society for Horticultural Science (Krizek and McFarlane, 1983) (see Appendix H). Although these guidelines were prepared for research in greenhouses and growth chambers, they should apply equally well to intensive field studies.

6. Data Acquisition

Data acquisition is either manual through use of recorders and notes, or automated through use of a datalogger or interface/computer system. Manual data acquisition is labor intensive and conservative in terms of retention of all continuous data traces. However, reduction and averaging of data for desired periods, e.g., 1-hour ozone concentrations, is subjective for each worker.

Automated systems with dataloggers facilitate storage of large amounts of data from a variety of monitors in a form amenable to computer processing and reduction. Computerized systems using an instrument interface and microcomputer allow for on-line storing, processing, and retrieval of pollutant and environmental data. Programs have been developed at several sites to present a continuous printout of both summary tabular and graphic data.

Use of a computerized system further facilitates control of pollutant dispensing via signal feedback to ozone generators, solenoids, critical orifices, flow controllers, or other devices. The pollutant dispensing can be controlled to provide specified exposure profiles ranging from continuous constant added concentrations to proportional increases based on ambient concentrations.

Automated data acquisition systems generally are programmed to save only average concentrations for longer periods of time, typically 1 hour. This permits storage of continuous data for longer periods than if short averaging times, i.e., minutes, are used. However, peak concentrations of short duration are masked by longer averages, even though peak concentrations may be biologically significant. More sophisticated acquisition systems may be able to retain both average and peak concentrations.

B. Wet Deposition Systems

Irrespective of the meteorological form of wet deposition or the location of the facility in a controlled or field environment, most systems have common structural components providing for water purification and conditioning, simulant stock solution storage, simulant mixing and flow control, droplet generation and dispensing, presentation/display of vegetation to the hydrometeor, and characterization of the deposition rate and hydrometeor chemistry. The uniqueness of the facilities for simulating rainfall, mist/cloud water, or aerosols lies in the selection of methods for droplet generation and dispensing; most of the other components are common to all systems. Consequently, the facilities will be discussed according to the form of the hydrometeor being generated. The concerns of the degree of environmental modification and control resulting from the exposure system employed is the same as for dry deposition exposure facilities. More detailed descriptions of these facilities are provided in Appendix F. The issue of snowfall deposition is not addressed in this document.

1. Rainfall Simulation Indoors

Irrespective of whether rainfall simulation is conducted in a controlled environment or under field conditions, only two methods are commonly used. In the first, rainfall is injected into the atmosphere via a nozzle that disassociates the liquid into a range of droplet sizes depending on the configuration of the nozzle and pressure of the feed solution. Typically, these systems have one or several nozzles arranged to deliver droplets in a uniform pattern. Many controlled environment systems use multiple nozzles of similar or different configuration to provide either more complete coverage and/or a range of droplet sizes within a single event. There are alternative methods for droplet generation (e.g., fixed orifice size) which are discussed in the section on pollutant dispensing and control. None of the rainfall simulators in controlled environments use enclosures, so that the environment reflects that of the glasshouse or growth chamber.

2. Rainfall Simulation Outdoors

While the most common method of generating rain in field situations is identical to that in controlled environments, i.e., droplet generation via nozzle, the uniqueness of the different types of field exposure systems lies in their methods of addressing incident ambient rain and the protocol for the distribution of simulated rain. The use of enclosures (shelters), either continuously or intermittently, may inadvertently modify the plant's microclimate either during the exposure event itself or subsequently (i.e., non-exposure environment). Techniques that apply simulated rain in excess of that provided naturally do not suffer from the same problems as those induced by enclosures, but, alternatively, they must evaluate the influence of additional water and chemical inputs on the system's microclimate and edaphic properties. Techniques that apply simulated acid rain in excess of that provided naturally do suffer from the inherent problem of uniform distribution of applied simulated rains, particularly in windy climates. This may prove to be a major limitation at high elevations where high winds are common.

Rainfall exclusion is commonly achieved using either permanently-mounted covers or movable exclusion lids that are activated with an electronic rain sensor. Permanent exclusion systems create environments that have many properties comparable to those of a glasshouse (except for the soil component), and the potential for climate modification must be realized. The method of automated exclusion uses lids over either small individual plots (e.g., 7.5 m²) or large surface areas (e.g., 300 m²). Most field systems that exclude ambient rain (either automatically or permanently) dispense simulated rain on a prescheduled basis (scheduled addition) rather than simultaneously with the event and under the same environmental conditions (automated addition). The most common protocol calls for scheduled additions either early in the morning (0600-0900 h), at night, or on cloudy days. Field exposure systems are documented that provide automated methods of exclusion and application in a realistic and simultaneous fashion with respect to ambient rainfall events. The selection of automated versus scheduled additions of rainfall should be based on the experimental objectives. Scheduled additions may provide greater control in evaluating the effects of wet deposition on terrestrial vegetation.

Methods of simulating rainfall in excess of ambient rain (shelterless plots) are many and include both small (e.g., 7 m²) and large (e.g., 625 m²) plot simulations. The techniques are applicable to both forested and agricultural landscapes. Droplet generation and dispensing are achieved by either a system of distributed nozzles or a rotating boom that distributes rain droplets along a radial arm that circulates around the plot providing rain at intermittent intervals.

3. Mist/Cloud Water Simulation

The most common method for delivering wet deposition in mist/cloud water form is similar to that for rainfall and uses specific nozzles or rotating discs to create droplets of the size common to cloud water (5-50 µm dia.). Unlike rainfall systems, most cloud water exposure systems require enclosures to achieve atmospheric hydrometeor concentrations and/or deposition rates that realistically mimic ambient. The enclosure size is limited by the ability of

the dispensing system to saturate the atmosphere. The use of enclosures may significantly modify the plants' microclimate in a fashion similar to that observed for rainfall systems. The issue is less acute if the enclosures are used only during cloud water events. One common problem in cloud water systems is the absence of a method for generating the inertial impaction necessary to simulate realistic deposition/interception rates by canopies. Some special aspects of the cloud water/plant interactions can be addressed using wind tunnels, notably the biological (e.g., canopy structure) and physical (e.g., wind speed) factors governing foliar interception of cloud droplets. It is important in mist/cloud water systems to characterize deposition rates to various components of the canopy.

4. Aerosol Simulation Systems

The method of simulating aerosols requires droplet sizes $< 1.0 \mu\text{m}$ in diameter and a well controlled environment with respect to temperature and vapor pressure. Unlike mist/cloud water exposure systems, high wind speeds are not required to generate realistic deposition rates. Aerosol generation can be achieved using specifically designed impingement nozzles. Methods of characterizing the atmospheric chemistry and deposition differ significantly from those used for simulators of rainfall and cloud water. As with cloud water systems, it is important to characterize precisely both atmospheric chemistry and foliar deposition rates if the system is to provide useful information regarding the response of forest species.

5. Simulation of Both Wet and Dry Deposition

Only a few systems provide automated control of both wet and dry deposition; examples are available for both controlled environments and field conditions. The most common field systems use: (1) a zonal air pollution exposure system in combination with rainfall simulation without excluding natural rain (i.e., shelterless plot); (2) open-top chambers amended with automated systems for rainfall exclusion and real-time simulation; and (3) forced air exclusion of ambient gaseous air pollution (via duct system) combined with exclusion of natural rain and subsequent real-time rainfall simulation.

6. Pollutant Monitoring

The methodology for monitoring wet deposition pollutants must recognize the need to estimate both the rate of hydrometeor deposition and interception, as well as the chemical composition of the hydrometeor. While the methods for characterizing deposition are hydrometeor-dependent (i.e., function of hydrometeor size and mass), the methods for chemical analysis are generally common among all forms of wet deposition and involve standard analytical procedures. The methods of characterizing hydrometeor deposition will be discussed first.

With respect to rainfall collection either in the field or in controlled environments, the most common method used in wet deposition research is to assume a uniform distribution of simulant volume across the surface area receiving rain so that deposition rates are inferred rather than measured. In those systems in which deposition was actually measured, the coefficient of variation ranged from 5-20% in systems with rotating platforms (e.g., Chevone

et al., 1984). The degree of variation of hydrometeor deposition in systems using fixed platforms will be at least as great. The recommended method of collecting rainfall is analogous to that used in field settings (i.e., a wet-only collector) particularly if the collection will subsequently be analyzed chemically. The most suitable container is plastic (polyethylene), positioned to collect incident rain simultaneously with exposure of vegetation (Shriner, 1972).

The monitoring methods used in studies of mist/cloud water deposition are not well documented. Many systems distribute mist via a manual dispenser "uniformly" over all vegetation and calculate by inference a deposition rate in mm (per leaf area). Empirical approaches in simulation studies are encouraged to include: (1) collection of both incident and throughfall components; and (2) gravimetric analysis of vegetation before and after cloud water events with subsequent deposition rates calculated for foliar surface area. Under field conditions, the collection method depends on the advective component of the precipitation event. In areas where inertial impaction is the major factor governing hydrologic input (e.g., high elevation forests immersed in clouds), a passive water collector can be used (Lovett et al., 1982), in which cloud water is impacted on lengths of monofilament nylon line strung sloping in a plastic frame. The impacted droplets coalesce, run down the line, and are collected in a bottle at the base for subsequent chemical analysis. Given the collection rate per surface area of line, wind speed, and capture efficiency of the line, the cloud's liquid water content can be estimated. Lovett (1984) estimated that the uncertainty in the collection efficiencies of this technique is $\pm 25\%$.

Active methods of collecting cloud water/mist are more commonly used in radiation fogs and mists in which deposition to tree species is not governed by inertial impaction. Using forced air (either positive or negative pressure), a defined rate (l/min) of mist-laden air is impacted on teflon line or a tube; the droplets coalesce, run down the line, and are collected in a bottle for subsequent analysis (e.g., Global Geochemistry Corporation, Fog Samples). These systems are also subject to error, most likely related to the less than complete collection of large droplets.

Suspended aerosols/particles are commonly collected by a combination of three techniques: filtration, impaction, and impingement, with most field applications using filtration and impaction. With due consideration to collector design, the analysis can focus on the particle size distribution of the aerosol/particles. The problems associated with collecting aerosols are not trivial and in general are a greater source of error than that associated with the chemical analysis (Lindberg and McLaughlin, 1986). The same conclusion should be extended to both rainfall and cloud water/mist.

Irrespective of the form of wet deposition, the following assumptions should be empirically evaluated for any laboratory or field-based exposure system:

1. inferred deposition rates (calculated based on simulant volume dispensed and platform surface area) are accurate;
2. distribution of deposited hydrometeor is uniform in space and time;

3. collector efficiency (particularly important for mist/cloud water and aerosols).

In practice, few systems satisfy these requirements.

An equally important parameter to characterize in pollutant monitoring is the site of hydrometeor deposition within the canopy. The observation that nearly all of the deposition in the form of mist/cloud water is in the upper 50% of the forest canopy in the high elevation forests of North America (Lovett, 1984) indicates that deposition is not uniform throughout the canopy. Current field techniques for characterizing patterns of within-canopy deposition rely on direct measurement of throughfall and stemflow during cloud water immersion coupled with fog droplet interception models that estimate sites of deposition in the canopy depending on leaf area index, wind speed, and droplet size (Lovett, 1984). In a controlled environment setting, cloud water deposition can be estimated by throughfall analysis (e.g., Reiners and Olson, 1984) or gravimetric techniques. For aerosol deposition, gravimetric techniques are insensitive, and one must resort to a tracer tagging of the aerosol and subsequent elution from the foliage (e.g., Thorne et al., 1982).

The chemical analysis of incident hydrometeors is a function of the experimental design. The relevant classes of components in precipitation for consideration include pH, conductivity, multiple inorganic cations (H^+ , Ca^{+2} , Mg^{+2} , Na^+ , K^+ , NH_4^+ , Al^{+3} , Fe^{+3} , and Mn^{+2}) and anions (SO_4^{-2} , NO_3^- , Cl^- , HCO_3^- , and PO_4^{-3}), dissolved trace metals (Pb, Zn, Cu, Ni, Cd, and Cr), reactive chemical species (H_2O_2), organic acids (Shriner, 1979), and solids (Unsworth, personal communication). Several individual components may require on-site/rapid analysis prior to preservation (e.g., H_2O_2 , NH_4^+ , pH). The degree of variation in the chemistry of simulated rain is not well characterized in space or time for any type of exposure system (batch or in-line mixing). In general, most systems rely on select indices of simulant chemistry (e.g., pH, conductivity) to characterize composition. In fact, only a few facilities routinely analyze the rain simulant; most assume the composition is accurately reflected in the recipe.

7. Pollutant Dispensing and Control

The critical features with respect to physically and chemically simulating wet deposition in the form of rainfall, mist/cloud water, and aerosols are the following:

- a. chemical composition of the hydrometeor;
- b. spatial distribution of the hydrometeor;
- c. event intensity or rate (cm/h);
- d. event duration (h);
- e. frequency of events (e.g., events/wk, if events occur at regular intervals);

- f. temporal dynamics of events relative to intermittent periods of dry deposition if events are distributed in either a stochastic or time-series fashion;
- g. distribution of hydrometeor sizes (mm dia);
- h. terminal velocity of hydrometeor (m/s); and
- i. changes in chemical composition within and among events.

The components involved in hydrometeor dispensing and control in wet deposition systems commonly focus on the hydrometeor itself rather than the chemical species in solution. The conditioning of makeup water is typically achieved using deionization and/or distillation of process or tap water. According to a recipe of desired chemical composition, stock solutions are mixed in concentrated form and subsequently stored. Dilution is achieved manually or through an automated mixing and control system that proportions the volume of deionized water relative to that of the concentrated simulant. This mixing and dilution is achieved either by the batch or in-line mode, the latter requiring a higher degree of quality assurance. The actual dispensing is achieved through nozzle injection or droplet release from an orifice (e.g., a distributed array of needle tips). To minimize non-uniform distribution of the hydrometeor, many systems place the plants on a rotating platform.

Both dispensing techniques (nozzles and needle tips) operate under positive pressure and are flexible enough to accommodate most of the important features of rainfall simulation. Nozzles disassociate the simulant solution into droplets, the size of which depends on the nozzle specification and feed solution pressure. Most systems inject droplets upward, allowing the hydrometeors to fall by gravity. This technique facilitates a realistic terminal velocity of the droplet and usually promotes a more uniform distribution. The method of release of simulant under pressure through a series of openings along a boom or hub has been successfully used in both field (e.g., Abrahamsen et al., 1977) and laboratory (e.g., Chevone et al., 1984) studies. The selection of needle tips with defined small openings can effectively control droplet diameter, although the tips must be geometrically distributed to achieve uniform dispensing along the length of the boom due to in-line pressure drops.

In either case, control of dispensing rate is provided through the regulation of flow rate and pressure. Programmable variable-speed motors can be used in conjunction with flow controllers to regulate the intensity of rain and intermittent frequency of events. Under field and laboratory conditions, most systems dispense simulants in a pre-scheduled protocol providing a fixed/constant duration of events and intervening dry periods. Control or computer-assisted systems for field application have been documented to simulate rainfall deposition to shielded plots on a real-time basis by providing on-line feedback to match deposition rates between ambient and simulated plots (e.g., Johnston et al., 1986). This matching is provided by a tipping bucket rain gauge that signals a control whenever the rain increment exceeds a set point.

8. Environment Monitoring

The degree of environmental monitoring in wet deposition studies is usually limited to the evaluation of light, temperature, and photoperiod in controlled environments and light, temperature, and gaseous pollutant level (i.e., ozone) in field systems. The majority of monitoring data is reported as a single season mean value without a corresponding dispersion statistic or analysis of temporal dynamics. There are notable exceptions to this practice depending on the individual researcher's interest. Efforts to characterize those environmental factors governing the residence time of droplets on the leaf surface (i.e., temperature, vapor pressure, light) should be a high priority.

9. Data Acquisition

Few systems include data acquisition features, and the only documented examples are those from field exposure systems using either open-top chambers (e.g., Johnston et al., 1986) or track exclusion systems (Kuja et al., 1985). These field systems have acquisition capabilities comparable to those described in the section on field exposure systems for dry deposition research. The only unique features relate to the wet deposition environment and include records of the ambient rainfall rates and times, corresponding deposition data in simulated plots, duration and frequency of rainfall events, simulant dispensing rate, and the status of the various reservoirs for storage of stock solutions. It is suggested that acquisition capabilities be extended to characterize soil water potential, leaf surface wetness, and some technique for monitoring simulated rainfall distribution and chemistry in treated plots. Most systems use a personal computer (e.g., Apple IIe, IBM AT) with either "off the shelf" or personalized software packages.

III. Criteria for Evaluating Exposure System Performance

Irrespective of the deposition mode (i.e., wet or dry), the facility location (i.e., outdoors or indoors), and/or the study objective, generic criteria can be specified to evaluate the performance of any exposure system. Criteria were identified which convey in a quantitative sense the "realism" in exposure systems with regard to chemical and physical features of the atmosphere (including the pollutant), physiological and growth processes of the plant, and physical and chemical properties of the soil. The criteria are listed in Table III-1 and represent aspects that should be considered from the widest point of view. The intent is to assure that the exposure systems: (1) select and characterize the pollutant exposure regime in the atmosphere with respect to those occurring under ambient conditions (in both a chemical and a temporal fashion) or in a desired fashion; (2) select and characterize the environment (during exposure and non-exposure periods) with respect to the ecological and physiological requirements of the species; and (3) provide an environment that generates pollutant deposition rates and patterns that can be compared to those occurring under ambient conditions in agricultural or forested landscapes. The relative importance of the various criteria will depend on the objective of the research task and the capabilities of the exposure system. Consequently, the criteria should not be applied indiscriminantly to all systems. Moreover, the selection of relevant criteria for even a single exposure system will differ as a function of experiment-specific objectives.

The criteria for evaluating a system's performance fall into three general categories: (1) pollutant chemistry in the atmosphere; (2) the edaphic, climatic, and atmospheric environment both during exposure and non-exposure periods; and (3) the biological attributes of the plant.

A. Pollutant Chemistry in the Atmosphere

The principal concerns with dry deposition facilities relate to the chemical's concentration on both a spatial and temporal scale (Table III-1). Within a given exposure system, it is important to characterize the distribution of the pollutant within the canopy. While some systems foster instantaneous mixing throughout the canopy (e.g., CSTR), others promote pollutant gradients in either a downward (down-draft chambers) or an upward (open-top chambers) direction. Depending on the degree of turbulence and canopy closure (either within or among chambers), pollutant distribution on a horizontal plane may vary significantly. With respect to temporal exposure patterns, it is important to characterize the gas concentration dynamics as a function of the exposure period relative to the plant's daily physiological functions (e.g., stomatal conductance) and seasonal phenology. These features include time of exposure relative to the time of photoperiod initiation and the growth stage of the plant. Equally important are characterizations of the duration of exposure and the stochastic nature of exposure events and intervening respite periods. This criterion may require statistical characterizations that convey the episodic/temporal dynamics of exposure patterns rather than simply a mean concentration.

Table III-1. Potentially relevant criteria to use in evaluating exposure system performance. Criteria will vary depending on deposition mode and experiment design.

A. Chemical Properties of the Pollutant in the Atmosphere

1. Dry Deposition

a. Spatial Features

(1) Concentration Distribution (Uniform versus Non-Uniform)

(a) Vertical Plane (e.g., Profiles)

(b) Horizontal Plane (e.g., Patches)

b. Temporal Features

(1) Exposure Profiles

(a) Square Wave

(b) Time Series

(2) Daily Exposure Regimes

(a) Diurnal

(b) Nocturnal

(3) Seasonal Exposure Regimes

(4) Annual Exposure Regimes

(a) Growing Season

(b) Winter Season

(5) Episodes

(a) Episode Duration

(b) Respite Duration

(c) Stochasticity/Periodicity of Events

2. Wet Deposition

a. Solution Chemistry (Pre-Droplet Formation)

(1) Cations

(2) Anions

(3) Organics

(4) Reactive Chemical Species

(5) Acidity/pH

(6) Conductivity

b. Incident Hydrometeor Chemistry [same chemical species as (a) above]

(1) Spatial Features

(2) Temporal Features

(a) Exposure Profiles

(b) Daily Exposure Regimes

(c) Seasonal Exposure Regimes

(d) Annual Exposure Regimes

(e) Episodes

c. Incident Hydrometeor Physics

(1) Spatial Features

(a) Horizontal-Plane Distribution

(b) Vertical-Plane Distribution

(2) Temporal Features

(a) Rate

(b) Duration

(continued)

Table III-1 (continued)

-
- (c) Frequency
 - (d) Time of Day/Season
 - (e) Inter-Event Features
 - d. Incident Hydrometeor Physical Features
 - (1) Droplet Size Statistics
 - (2) Terminal Velocity
 - (3) Chemical-Specific Deposition Rates
 - (a) Spatial Distribution
 - (b) Temporal Distribution
 - (1) Event
 - (2) Seasonal
 - (3) Annual
 - 3. Combination of Pollutants (Dry/Dry, Wet/Wet, Wet/Dry)
 - a. Characteristics of Co-Occurrence
 - b. Characteristics of Independence
 - c. Relationship Between Wet Deposition Event and Antecedent Dry Deposition Period
- B. Physical and Chemical Features of the Environment
- 1. Atmosphere
 - a. Radiation
 - (1) Spectral Quality, Including the Ultraviolet Component (PAR)
 - (2) Quantity
 - b. Temperature/Moisture
 - (1) Air Temperature
 - (2) Leaf Temperature
 - (3) Leaf-to-Air Differentials (vapor pressure deficit)
 - (4) Dew and Frost Formation
 - (5) Thermoperiod
 - c. Turbulence and Mixing
 - (1) Canopy Aerodynamic Resistance to Turbulent Transfer
 - (2) Leaf Boundary Layer Resistance to H₂O
 - (3) Wind Speed
 - d. Turnover Time of Air Reservoir
 - e. Ambient Hydrometeor Deposition
 - f. Trace Gases
 - (1) Carbon Dioxide
 - (2) Ethylene
 - (3) Water Vapor
 - 2. Soil
 - a. Chemistry
 - (1) Cation Exchange Capacity
 - (2) Partial Pressure of Oxygen
 - (3) Soil Solution Chemistry
 - (4) Soil Nutrient Analysis
-

(continued)

Table III-1 (continued)

-
- b. Physics
 - (1) Temperature
 - (2) Water Potential
 - (3) Porosity/Solution Infiltration
 - C. Biological Features of the Environment
 - 1. Vegetation
 - a. Structure
 - (1) Leaf Area/Leaf Area Index
 - (2) Canopy Architecture
 - (3) Leaf Developmental Stage/Phenology
 - (4) Canopy Height Relative to Chamber
 - (5) Canopy Girth Relative to Chamber
 - (6) Belowground Root Architecture
 - (7) Mycorrhizae
 - b. Function
 - (1) Net Photosynthesis
 - (2) Transpiration
 - (3) Leaf Water Potential
 - (4) Leaf Conductance to H₂O
 - (5) Growth Rates
 - (6) Biomass Partitioning Above- and Belowground
 - (7) Chlorophyll Content
 - 2. Deposition of Pollutants
 - a. Wet Deposition
 - (1) Foliar Interception of Hydrometeor
 - (a) Horizontal-Plane Variation
 - (b) Vertical-Plane Variation
 - (2) Foliar Retention of Hydrometeor
 - (3) Chemical Processing of Intercepted Hydrometeor
 - (a) Throughfall Chemistry
 - (b) Stemflow Chemistry
 - (4) Deposition to Soil Surface
 - (5) Chemical Processing of Hydrometeor Chemicals in Soil/Litter
 - b. Dry Deposition Rates and Amounts
 - (1) Deposition to Individual Leaves
 - (a) Leaf Surface
 - (1) Dry Surface
 - (2) Wet Surface
 - (b) Leaf Interior
 - (2) Deposition to Canopy
 - (3) Deposition to Chamber Walls/Surfaces
 - (a) Wet
 - (b) Dry
 - (4) Deposition to Soil/Litter Surface

(continued)

Table III-1 (continued)

D. Hardware

1. Dry Deposition

a. Chamber Air Handling

- (1) Air Exchange Rate -- Turnover Time
- (2) Air Movement -- Recirculation, Single-Pass
- (3) Equilibration Time
- (4) Direction of Air Flow
- (5) Air Pressure
- (6) Air Leakage
- (7) Air Filtration

b. Chamber Characteristics

- (1) Pollutant Sorption
- (2) Size
- (3) Volume-to-Leaf Area Ratio

c. Pollutant Concentration

d. Environment

- (1) Monitoring
- (2) Control

e. Data Acquisition

2. Wet Deposition

a. Hydrometeor Generation

- (1) Makeup Water Conditioning
- (2) Generation of Droplets
- (3) Distribution/Dispensing
- (4) Monitoring/Control

b. Environment

- (1) Monitoring
- (2) Control

c. Data Acquisition

For wet deposition systems, criteria should address the physical form and chemical composition of the incident hydrometeor (Table III-1). It is important to recognize that analyses must be conducted on the incident hydrometeor -- rain, mist/cloud water, and aerosol -- in addition to the stock solution. Equally important is a rigorous analysis of deposition rate and, at least in some situations, an evaluation of the deposition site. This aspect is critically important in systems designed to simulate cloud water and aerosols. As with dry deposition, the event's temporal dynamics must be characterized with respect to duration, intensity, and frequency. Of lesser importance is an analysis of physical features characterizing droplet size and velocity. Depending on the experimental design, it may be necessary to evaluate the chemistry of the hydrometeor after deposition either on the leaf surface (droplet evaporation rate) or in throughfall/stemflow.

In those systems providing pollutant mixtures either in the form of gases or gases plus wet deposition, not only must the individual pollutant regimes be characterized, but their temporal relationships evaluated with respect to the ambient environment. This requires attention focused on exposure protocols that convey the characteristics of pollutant co-occurrence, characteristics of pollutant independence, the relationship between wet deposition events and antecedent periods of dry deposition, and the phenology of the plant.

B. Physical and Chemical Features of the Environment

These criteria address the climatic, edaphic, and atmospheric environment during both exposure and non-exposure periods (Table III-1). It is critically important to recognize the potential for inadvertent modification of the plant's microenvironment by the exposure system such that the plant's response to the air pollutant is not typical nor is its interaction with other biotic (e.g., pathogens) and abiotic (e.g., drought, winter temperature) stresses. Attention must focus on both the aboveground and belowground environment of the plant.

Most exposure systems influence both physical and chemical properties of the atmospheric environment; the degree of modification varies not only among facilities of dissimilar design but also within a given facility as a function of time of year, time of day or night, and canopy architecture and biomass. The most relevant physical properties include: (1) the radiation balance (spectral quality and quantity of incident light) and photoperiod; (2) heat and moisture relationships (e.g., thermal gradients, leaf/air temperature, dew and frost formation, vapor pressure deficit); (3) turbulence and mixing of the air reservoir which affects the canopy's aerodynamic resistance to turbulent transfer, latent energy exchange, individual leaf boundary layer resistance to diffusion of gases, and residence/turnover time of the air reservoir; (4) wind speed; and (5) normal wet deposition processes as rain or cloud water. It must be recognized that these changes in a plant's microclimate are due to imposition of an exposure system and may be perpetuated even after the system is removed.

The potential changes in the chemical properties of the atmosphere are equally important. Exposure systems may inadvertently influence the concentration and distribution of physiologically-important gases such as carbon

dioxide, water vapor, and trace contaminants. Depending on the quality of the source materials used to generate pollutants or provide enclosures, as well as the residence time of the air reservoir in the enclosure, plants may inadvertently experience elevated levels of trace gases (e.g., ethylene) that affect physiological and growth processes (Morison and Gifford, 1984). Regarding wet deposition facilities specifically, the deposition of rain in excess of ambient may significantly affect the systems' performance. The physical and chemical modifications of the environment with excess wet deposition warrant evaluation with particular attention focusing on leaf surface wetness and edaphic properties, including soil water potential, partial pressure of oxygen, and soil solution chemistry. The use of lysimeters for soil water sampling at various depths in the profile is strongly encouraged.

C. Biological Attributes of the Plant

Biological (plant) factors are of great importance in the selection process for a suitable exposure system (Table III-1). The biological objectives (e.g., strain testing, physiological studies, dose-response determinations) may dictate the precision required in the research facility. The nature of the plants themselves will determine such aspects as size of facility and may affect the levels of both chemical and physical factors of the experimental environment.

This is particularly relevant to woody plant species whose growth characteristics (i.e., growth rate, periodicity in the initiation/elongation of new foliage, the substantial photosynthate allocation to structural and storage tissues in the aboveground and belowground tissues) render insensitive many of the more common indicators used in agricultural settings (e.g., degree of canopy closure, leaf/plant stage of development, anthesis, podfill). If the biological status of the plant differs markedly from that in more natural landscapes, the data are of less merit in efforts to predict how woody plants will respond to air pollution stress under field conditions. It is entirely possible that, while the exposure system may meet the established criteria for physical and chemical performance, it may be unacceptable based on the biological criteria.

The selection of biological criteria focuses on two aspects of plant/environment interactions: the rate of growth and associated physiological activity in both the root and shoot, and the foliar uptake of trace gases or interception of hydrometeors on vegetation surfaces (Table III-1). Examples exist in the published literature in which: (1) the physiological/morphological factors governing deposition fostered abnormally high or low pollutant fluxes; or (2) the rate of carbon gain in control plants only slightly exceeded the allocation of photosynthate to maintenance costs (i.e., no net growth). In either case, the presence or absence of statistically significant effects becomes biologically irrelevant.

The functional criteria with which to evaluate a system's and a plant's performance with respect to biological features include photosynthesis, respiration, transpiration, leaf water potential, components of leaf conductance to water vapor, and chlorophyll content. Foliar gas-exchange rates should be normalized to projected leaf area for broadleaf species and needle dry

weight or area for conifers (i.e., rates for performance evaluation purposes should not be reported on a per plant basis). The evaluation should be species- and environment-specific, using as a guideline values that are within $\pm 10\%$ of the corresponding rate in open air/companion plots. In controlled indoor exposure systems, rates of net photosynthesis and transpiration should be $> 70\%$ of those reported in more natural settings (Table III-2). These criteria assume that any influence of the exposure environment on foliar gas-exchange processes causes a proportional change in the allocation of photosynthate to structure, storage, and maintenance sinks. If that assumption is not realized, the applicability of the data is questionable.

Leaf conductance to water vapor is a measure of foliar porosity to the diffusion of gases. Boundary layer conductance should be $> 200 \mu\text{mol}/\text{cm}^2/\text{s}^1$ (i.e., ca. $0.2 \text{ s}/\text{cm}$ resistance), while stomatal conductance will vary as a function of species, environmental conditions, and phenology. Reasonable values under optimum growing conditions in the light range from $4\text{--}20 \mu\text{mol}/\text{cm}^2/\text{s}$ for broadleaf species and $1\text{--}4 \mu\text{mol}/\text{cm}^2/\text{s}$ for conifers (Table III-2). Substantially higher conductances to water vapor are characteristic of herbaceous species grown under optimal conditions (Table III-2).

Leaf water potential is a direct measure of the plant's water relations and is particularly important in exposure systems that either maximize canopy/leaf ventilation rates (e.g., open-top chambers, forced air exclusion systems) or simulate wet deposition. Estimates of leaf/twig/needle water potential should characterize both predawn and midday potentials at intervals throughout the season with more frequent sampling during periods of low soil water availability. Chlorophyll content may relate to photosynthetic capacity and the progression of foliar senescence and chlorosis/necrosis, and thus this criterion may serve as an indicator of plant vigor.

In woody plants, the selection of features to characterize growth rate should focus not only on leaf/needle initiation and elongation/expansion, but the allocation of photosynthate to structure and storage tissues. Reasonable rates of dry matter accumulation normalized for leaf area per day are $0.1\text{--}0.2 \mu\text{g}/\text{cm}^2/\text{d}$ for woody species and an order of magnitude higher for herbaceous species (Table III-2). Attempts should be made to assure that the chamber environment does not significantly alter the allocation patterns. This should include estimates of biomass in shoot and root tissues, with the latter further partitioned into large and fine root fractions.

Structural features are particularly important in woody plants, whose growth rates and architecture are far more variable between and among species than those of the more genetically uniform, domesticated crop species. Important aboveground criteria to consider include leaf area/leaf area index (LAI), canopy architecture and height (relative to the chamber's dimensions), and canopy girth (relative to the chamber's dimensions). Because the LAI affects pollutant deposition/interception rates as well as the chemistry of throughfall and stemflow, this criterion is particularly important in wet deposition studies under field conditions. Canopy architecture in general and height specifically may significantly limit the utility of some chamber systems, because many systems for dry deposition research in the field cannot adequately restrict the incursion of turbulent eddies into the chamber's air

Table III-2. Reasonable rates for select biological criteria as reported under near optimum growing conditions (Larcher, 1980; Korner et al., 1979).

Criteria	<u>C₃ Agricultural Crops</u>	<u>Woody Plants</u>	
	C ₃ Plants	Broadleaf	Conifer
Net Photosynthesis			
($\mu\text{mol}/\text{cm}^2/\text{h}$)	4.4-8.8	2.2-4.4	0.9-3.3
($\mu\text{mol}/\text{g}/\text{h}$)	---	---	68-410
Transpiration			
($\text{mmol}/\text{cm}^2/\text{h}$)	1.9-2.2	0.4-0.9	0.5-0.6
Leaf Conductance to H ₂ O			
($\mu\text{mol}/\text{cm}^2/\text{s}$)			
Boundary Layer	80-400	80-400	---
Stomate	20-130	4-20	1-4
Growth Rate			
($\mu\text{g}/\text{cm}^2/\text{d}$)	1.0-2.5	0.1-0.2	---

reservoir. Moreover, most wet deposition systems are designed to deliver droplets in a distributional uniformity that decreases with proximity to the nozzle.

Belowground criteria include root architecture, mycorrhizal infection and colonization. Root architecture is particularly important in stress interaction studies addressing the role of drought or soil nutrient status because species and even half-sib families differ substantially in their ability to explore and extract resources (water and nutrients) from different horizons of the soil profile. While the significance of mycorrhizae in resource acquisition in the soil (nutrients and water) and trace metal toxicity is recognized in woody species, and qualitative and quantitative analysis of mycorrhizal abundance should be a deliberate criterion of performance in exposure systems, it should not be forgotten that the existence of desirable mycorrhizal flora in the rhizosphere dictates that a significant fraction of the total carbon gain (as much as 20%) will be diverted away from other sinks to the fine roots.

IV. Evaluations

A. Dry Deposition -- Gaseous Exposures

Gaseous dry deposition systems in the field can be categorized into groups and subgroups based on degree of environmental modification and configuration of the system.

1. Non-Chamber Plume Systems

Plume systems can be grouped within three categories: grid configuration (de Cormis et al., 1975; Lee et al., 1975; Runeckles et al., 1981); encircling systems where the entire area is surrounded by emitters (Greenwood et al., 1982; McLeod et al., 1985; Roberts, M., personal communication); and linear configuration (Mittler et al., 1980; Moser et al., 1980; Northrop, 1983; Thompson et al., 1984).

Two generic advantages of the plume systems are: (1) a lack of environmental modifications caused by hardware; and (2) pollutants are dispersed through the plant canopy the same as in ambient air. The systems result in variable temporal distribution of pollutant concentrations. General disadvantages are: (1) that no pollutant treatment is possible at concentrations lower than ambient, thus the effects of ambient pollutants cannot be specifically identified as they can be with air filtration methods; (2) dispersion of pollutants is completely dependent on ambient wind patterns and their temporal nature, resulting in episodic exposures not necessarily representative of ambient exposure conditions; and (3) exposure plots must be widely separated to prevent cross-contamination, resulting in practical difficulties in area requirements for large dose-response experiments.

The grid configured-type plume system has the advantages of: (1) relatively simple system of emitter tubes without a requirement for large blowers with concomitant high electrical power requirements; and (2) a uniform horizontal distribution of pollutants irregardless of wind direction. Vertical concentrations, however, are dependent on an appropriate distribution of emitters at different heights. Grid systems were originally developed without computer control of pollutant emissions, which required a wide dispersion of emitters over the plant canopy. The addition of computer control capabilities has allowed for much simpler arrays of emitters.

Encircling systems have the advantage of providing a uniform distribution of pollutant over the exposure area by releasing pollutants from any prevailing wind direction. The current circular and square systems are under computer control, allowing for rapid feedback control of concentrations, and for monitoring of pollutant concentrations at many points in the exposure area for precise determination of pollutant dose to the plant canopy. This type of exposure system provides a totally ambient pattern of pollutant transport over the plant canopy without any environmental modification. However, the system disadvantage is the requirement for a large surface area for pollutant dispersion. Vertical pollutant profiles have not been measured for most systems. A computer and instrument interface system is required for all exposures.

Linear configuration plume systems provide a range of pollutant concentrations over a relatively small space for regression analysis of pollutant exposure and plant response. However, an intensive array of pollutant sampling points is required to characterize the exposure adequately, and a sophisticated computer system is required for feedback control of the pollutant release. Linear plume systems are more dependent on wind direction for pollutant emission than encircling systems, resulting in more unpredictable pollutant concentrations and durations of exposure than the other plume configurations. This characteristic greatly reduces the flexibility of experimental designs and feasibility of experiment replication. The gradient of exposure concentrations also may be much greater than that appearing with ambient air, i.e., concentrations are much higher than ambient near the emitters.

2. Non-Chamber Air Exclusion Systems

Air exclusion systems can be grouped within two categories: duct systems (Jones et al., 1977; Kuja et al., 1985; Laurence et al., 1982; Shinn et al., 1977; Thompson and Olszyk, 1985); and the "gun" system (Spierings, 1967).

Air exclusion systems permit determination of the effects of ambient air on vegetation with the capability of adding pollutants to the plant canopy. Duct systems are midway between plume systems and chambers in terms of simplicity of design, construction, and operation. The ducts themselves are simple and inexpensive to construct and flexible in terms of length, hole direction, and row width. However, the blowers generally are of higher capacity than for chambers, resulting in increased construction and electrical costs. For relatively short plants, duct systems can provide effective exclusion or addition when inflated, with little modification of the plant environment. There are physical limitations to plant row width between the ducts, and a plant height limitation. Duct systems at present are suitable only for row crops. Air speed over the plant canopy is high enough to avoid problems with low leaf boundary layer conductances. The air flow pattern is below and across the plant canopy, and not necessarily representative of ambient conditions. Also, there is a large vertical gradient in pollutant concentrations. Concentrations decline rapidly above the ducts. There normally is not a large horizontal gradient in pollutant concentrations across the row, or down the row, unless the hole diameters are specifically designed to reduce or increase air flow along the length of the ducts.

In general, the environmental conditions between ducts are the same as in outside areas. However, during the winter, air and soil temperatures are generally greater between ducts than outside. Light intensity can be higher or lower between the ducts than outside, depending on the sun angle.

The "gun" air exclusion system was developed specifically to expose particular branches of trees to air pollutants. The "gun" has not been tested for pollutant uniformity and no information is available regarding its ability to exclude ambient air, or its effects on environmental conditions around the branch or gas exchange by the branch. The "gun" system is not adaptable for multiple tree exposures, but can give a carefully defined pollutant treatment to a specific target plant.

3. Chambers -- Outdoor

Field chambers can be grouped within three categories: open-top (Heagle et al., 1973; Mandl et al., 1973; Brewer, 1979, 1983; Ashmore et al., 1980; Buckenham et al., 1981; Brewer, 1983; Fowler D., personal communication; Heck et al., 1984; Roberts et al., 1983; Seelinger et al., 1985; Thompson and Olszyk, 1981); semi-open-top (Runeckles et al., 1978; Hogsett et al., 1985; Krause, G., personal communication, Olszyk, D., personal communication); and closed-top (Van Haut, 1972; Keller, 1976; Guderian, 1977; Roberts, 1981; Musselman et al., 1985b; Lucas P., personal communication).

The generic advantage of field chambers is the ability to exclude ambient pollutants, and to enclose an experimental area for an increased range of treatments at near-ambient environmental conditions. The advantage of having a filtered enclosure (i.e., chamber) for a treatment range is also the basis for the generic disadvantage, i.e., the accompanying environmental modification during plant exposures.

Open-top chambers provide for the least amount of environmental modification of any outdoor chamber, coupled with ambient air exclusion of up to 80%. However, even a small degree of chamber-induced environmental modification (e.g., increased temperature and decreased solar radiation) may have a significant effect on plant growth, and this modification may be greatest during the cooler months. In addition to temperature change, all open-top chambers exclude at least some ambient rain and some designs can have "rain-shadows" resulting in variation in rainfall patterns within the chamber. The air flow in most chambers is across and/or from under the plant canopy which produces a pattern of air movement atypical of field conditions. Only those chambers with modifications designed to force air down and over the canopy are fully representative of ambient conditions. Open-top chambers have relatively low variability in pollutant concentrations horizontally across chambers. Vertical variability also is low if the chambers have some type of frustrum or baffle at the top to limit ambient air incursion. Smaller open-top chambers with high air exchange rates have the least deviation from outside air temperature and relative humidity, coupled with a high air speed over the plant canopy.

Exclusion of ambient pollutants is dependent on the filtration system in place on the air-intake for the open chambers. Activated charcoal, commonly used with most field chamber blowers, does not effectively remove all ambient air pollutants. Activated-charcoal removes much of the O_3 , but little of the SO_2 , NO , or NO_2 from the air. Chemically treated materials have a much greater filtering efficiency for a variety of air pollutants (D. Fowlers, 1985, personal communication). Purafil[®], potassium permanganate impregnated alumina particles, removes much of the ambient NO_2 , SO_2 , and also NO via a chemical reaction of NO to NO_2 (D. Mandl, 1986, personal communication). Fresh Purafil[®] also will remove ethylene. However, Purafil[®] does not have a high efficiency for removing O_3 . Ideally, a series of filters should be used to remove different ambient air pollutants prior to injection of air into the chambers. However, some chemically treated filtering materials, e.g., brominated charcoal, are fairly corrosive, thus requiring more resistant and expensive materials for filter containers.

Most current open-top field chamber designs have been widely used for growing season studies with many crop species, but have diameters and heights which currently limit their use for larger plants. Larger chambers that appear to provide the necessary volume for a small tree's canopy, have more environmental modification and fewer air exchanges compared to smaller chambers. Larger chambers may be adequate only for a single or a few trees per chamber, necessitating numerous chambers for replication within pollutant treatments.

Semi-open-top chambers permit control of the plant canopy atmosphere in areas where environmental conditions preclude open-top chambers. In particular, the semi-open-top designs may be best for exclusion of ambient precipitation.

Closed-top chambers allow for total control of the atmosphere in the plant canopy. Unlike open-top chambers, ambient pollutant exclusion is unaffected by outside wind speed, and is generally greater than open-tops, but still not 100% efficient. Vertical and horizontal variability in pollutant concentrations is lower than with any other system for field studies. Closed top chambers generally have lower irradiances and greater variability in relative humidity compared to other chamber types.

A comparative performance evaluation of all groups of gaseous dry deposition systems in the field is presented in Tables IV-1 and IV-2.

4. Chambers -- Indoors

Most of the indoor exposure chambers used in gaseous dry deposition research can be categorized into four types: chambers in greenhouses (Berry, 1970; Heck et al., 1978; Hill et al., 1959; Lockyer et al., 1976; Piersol and Hanan, 1975; Posthumus, 1978); self-contained chambers (Adams, 1961; Aiga et al., 1984; Hill, 1967; Jensen and Bender, 1977; Menser and Heggstad, 1964; Oliva and Steubing, 1976; Payer et al., 1985; Wood et al., 1973); chambers within chambers (Cantwell, 1968; Heck et al., 1968; McLaughlin et al., 1976; Payer et al., 1985); and CSTR's (Heck et al., 1978; Rogers et al., 1977). They have many characteristics in common, but there are some variations. A performance evaluation of indoor exposure chambers is presented in Table IV-3. A partial quantitative assessment based on reported values or characteristics is presented in Table IV-4.

There are many versions of the chamber-within-greenhouse approach to indoor gaseous dry deposition research, varying from using entire greenhouses as treatment chambers to using sets of small chambers or CSTR's within greenhouses. This approach has the advantage of providing large chambers, if needed, and several chambers, if needed, for concurrent differential-dose experiments. Partial control of temperature and humidity is provided by the greenhouse structure and light can be augmented by artificial sources placed over the chambers. The air movement required to minimize environmental differences between chambers and greenhouses may be sufficient to minimize leaf boundary layer resistance and to minimize gradients of environment and pollutants within chambers. Natural cycles and levels of light spectrum and irradiance are provided. This may help meet the objectives of some experiments. The disadvantages of greenhouse chambers include the marked seasonal and diurnal

Table IV-1. Performance evaluation for field systems for gaseous dry deposition research.

Criteria	Plume			Air Exclusion			Outdoor Chamber		
	Grid	Encircled	Linear	Ducts	Gun	Open	Semi-Open	Open	Closed
A. Atmosphere Chemistry									
1. Gaseous Pollutant									
a. Concentration Distribution									
(1) Vertical	Y/N ^a	Y/N	Y/N	Y/N	Y/N	Y/N	Y/N	Y/N	Y/N
(2) Horizontal	Y/Y	Y/Y	Y/Y	Y/Y	N/N	Y/Y	Y/Y	Y/Y	Y/Y
b. Temporal Patterns/Dynamics									
(1) Exposure Regimes	Y/N	Y/Y	Y/Y	Y/Y	Y/N	Y/Y	Y/Y	Y/Y	Y/Y
(2) Capabilities	Y/N	Y/Y	Y/N	Y/Y	Y/N	Y/Y	Y/N	Y/Y	Y/Y
2. Multiple Pollutant Exposures									
3. Non-Pollutant Chemicals									
a. CO ₂	N/N	N/N	N/N	N/N	N/N	Y/Y	Y/N	Y/N	Y/Y
b. H ₂ O Vapor	N/N	N/N	N/N	N/Y	N/N	Y/Y	Y/N	Y/N	Y/Y
c. Trace Gases	N/N	N/N	N/N	N/N	N/N	Y/N	Y/N	Y/N	Y/Y
B. Physical Properties of Atmosphere									
1. Radiation	N/N	N/Y	N/N	Y/Y	N/N	Y/Y	Y/Y	Y/Y	Y/Y
2. Heat Energy	N/N	N/Y	N/Y	Y/Y	Y/N	Y/Y	Y/Y	Y/Y	Y/Y
3. Air Movement	Y/N	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/N	Y/N	Y/N
C. Soil Environment ^b									
D. Hardware									
1. Plume									
a. System Configuration	Y/N	Y/Y	Y/N	---	---	---	---	---	---
b. Background Pollutants	Y/Y	Y/Y	Y/Y	---	---	---	---	---	---
2. Air Exclusion									
a. Degree of Exclusion	---	---	---	Y/N	Y/N	---	---	---	---
3. Chambers									
a. Equilibration	---	---	---	---	---	Y/N	Y/N	Y/N	Y/Y
b. Degree of Exclusion	---	---	---	---	---	Y/N	Y/N	Y/N	Y/Y
4. Pollutant Control--Monitoring	Y/N	Y/Y	Y/Y	Y/N	Y/N	Y/N	Y/N	Y/N	Y/Y
5. Environmental Control--Monitoring	Y/N	Y/Y	Y/Y	Y/N	Y/N	Y/N	Y/N	Y/N	Y/Y
6. Data Acquisition	Y/N	Y/Y	Y/Y	Y/N	Y/N	Y/N	Y/N	Y/N	Y/Y

a Important/characterized, Y = Yes, N = No for this type of system.

b The soil environment is important, especially for plants planted in soil. The soil environment can also cause problems for aboveground pots, especially in chambers.

Table IV-2. Performance values for field systems for gaseous dry deposition research.^a

Criteria	Plume			Air Exclusion			Outdoor Chamber		
	Grid	Encircled	Linear	Duct	Gun	Open	Semi-Open	Closed	
Number of Different Systems Described	3	3	4	5	1	15	7	7	
A. Atmospheric Chemistry									
1. Gaseous Pollutant									
a. Conc. Distribution									
(1) Horizontal (%)	5-300	10-100	5-1500	10-2300 (length)	---	6-30	6-20	10	
(2) Vertical (%)	25	40	34	10-70	---	6-63	5-10	---	
b. Temporal Patterns (%)	100	20	50-100	20	---	3-10	3-30	---	
2. Non-Pollutant Chemicals									
a. H ₂ O Vapor	Amb. ^b	Amb.	Amb.	Amb.	---	-10-+3%	---	+ 10%	
(Relative Humidity)									
B. Physical Properties of Atmosphere ^c									
1. Irradiance	Amb.	Amb.	Amb.	0-6%	---	-5-50%	-7-10%	-10-25%	
2. Heat Energy	Amb.	Amb.	Amb.	+0-1.5°C	---	+0.5-4°C	+1-5°C	+0.5-5°C	
3. Air Movement									
a. Over Canopy (m s ⁻¹)	Amb.	Amb.	Amb.	1.0	---	0.24-3.0	---	0.5-0.9	
b. Extra Turbulence	Amb.	Amb.	Amb.	High	High	Mod.	Mod.	Low to Hi	
c. Soil Environment ^c	Amb.	Amb.	Amb.	+0-2°C	Amb.	+1-3°C	+1°C	-1 to +1°C	
D. Hardware									
1. Ambient Air Exclusion (%)	None	None	None	70-80	---	50-80	40-70	75-90	
2. Plant Growing Area	120-1200	400-2100	5-784	7-30	N/A	1-20	0.4-7	1-13	
Per System (m ²) ^d									
3. Air Exchanges	N/A	N/A	N/A	N/A	N/A	2-10	1-4	1-4	

^a Variation or range reported for one or more different systems.

^b Ambient.

^c Change versus outside environmental conditions.

^d Total growing area, sampling area is often smaller.

Table IV-3. Performance evaluation for indoor exposure chambers for gaseous dry deposition research.

Criteria	Chambers in Green- houses	Self- Contained Chambers	Chambers within Growth Rooms	CSTR's
A. Chemical Properties				
1. Concentration				
a. Vertical Distribution	Y/Y ^a	Y/Y	Y/Y	Y/Y
b. Horizontal Distribution	Y/N	Y/Y	Y/Y	Y/Y
c. Chamber Losses	Y/N	Y/N	Y/N	Y/Y
2. Temporal Patterns/Dynamics				
a. Exposure Regimes	Y/N	Y/N	Y/N	Y/N
b. Capabilities	Y/N	Y/N	Y/N	Y/N
3. Multiple Pollutant Exposures	Y/N	Y/N	Y/N	Y/N
4. Non-Pollutant Chemicals				
a. Carbon Dioxide	Y/N	Y/N	Y/N	Y/N
b. Water Vapor (Humidity)	Y/Y	Y/Y	Y/N	Y/N
c. Trace Gases	Y/N	Y/N	Y/N	Y/N
B. Physical Properties				
1. Radiation				
a. Spectrum	Y/N	Y/N	Y/N	Y/N
b. Qualitative Features	Y/N	Y/Y	Y/N	Y/Y
2. Heat Energy				
a. Air Temperatures	Y/Y	Y/Y	Y/N	Y/Y
b. Leaf Temperatures	Y/N	Y/N	Y/N	Y/N
c. Gradients	Y/Y	Y/Y	Y/Y	Y/N
d. Dew and Frost	Y/N	Y/N	Y/N	Y/N
3. Air Movement				
a. Turbulence (Mixing)	Y/N	Y/N	Y/N	Y/Y
b. Air Exchange Rate	Y/Y	Y/Y	Y/Y	Y/Y
c. Chamber Equilibration	Y/N	Y/N	Y/Y	Y/Y
d. Direction	Y/Y	Y/N	Y/N	Y/Y
e. Wind Speed	Y/N	Y/Y	Y/Y	Y/Y
f. Overall System	Y/Y	Y/N	Y/Y	Y/Y
g. Pressure	Y/Y	Y/Y	Y/Y	Y/Y
4. Soil Environment	Y/N	Y/N	Y/N	Y/N
5. Unit Size	Y/Y	Y/Y	Y/Y	Y/Y

^a Important/Characterized, Y = Yes, N = No for this type of system.

Table IV-4. Performance values for indoor exposure chambers for gaseous dry deposition research.

Criteria	Chambers in Greenhouses	Self-Contained Chambers	Chambers within Growth Rooms	CSTR's
Number of Different Chambers Described	6	7	5	2
A. Chemical Properties				
1. Concentration				
a. Vertical Distribution	14% ^a	+ 3-5%	+ 5%	~ 0%
b. Horizontal Distribution	---	± 3-5%	± 5%	~ 0%
4. Non-Pollutant Chemicals				
a. Water Vapor (Humidity)	+ 5%	1-10%	---	---
B. Physical Properties				
1. Radiation				
a. Quantitative Features	---	+ 10-15%	---	+ 3.6%
2. Heat Energy				
a. Air Temperatures	+0.38-3.3°C	+0.3-1.0°C	---	+0.5-2.8°C
b. Gradients	+ 1.1°C	+0.6-2.2°C	---	---
3. Air Movement				
a. Turbulence (Mixing)	1 ^b	0	0	2
b. Air Exchange Rate	30-120	53-138	15-120	Variable
c. Direction				
(1) Down	4 ^b	3	2	---
(2) Horizontal	1	3	2	---
(3) Up	1	1	0	---
d. Overall System				
(1) Recirculation	1	3	1	0
(2) Partial Recirculation	0	1	0	2
(3) Single Pass-Through	5	3	4	0
e. Pressure				
(1) Negative	1	3	2	2
(2) Neutral	1	2	0	0
(3) Positive	4	2	2	0
5. Unit Size (Dimensions)				
a. Smallest	61x61x92 cm	122x107x122 cm	152x107x122 cm	81x137 cm
b. Largest	11x2.8x2.5 m	3x2.6x2.3 m	10x10x2.5 m	152x180 cm

^a Variation or range of variation reported for one or more different chambers.

^b Numbers of different chambers with this characteristic.

light patterns, and the shading by superstructures. There are likely to be gradients of temperature and humidity; the air movement required to minimize these may be excessive for plants. Temperature and humidity will vary according to the greenhouse conditions. A lack of repeatability due to changing seasonal environments may interfere with replication of experiments. Experimental designs must consider replication in both time and space.

Self-contained chambers have the advantage of providing the controlled and programmed temperature, light, and humidity characteristics of plant growth chambers. The environmental conditions are independent of outdoor environment conditions. This ensures repeatability of experiments on a year-round basis. The air movement required for environment control may provide the mixing to minimize gradients of pollutants within the chamber and crop canopy as well as to minimize leaf boundary layer resistance. Fluxes of gases to the plants may be calculated in well-sealed chambers for which inlet and outlet air volumes and composition are known. The disadvantages of self-contained chambers include the limitation of one treatment per chamber which necessitates several separate chambers for experiments. Corrosion of components by pollutant gases will be a problem with self-contained chambers. Where recirculation of air is used to permit flux fate studies or to minimize air conditioning requirements, there may be some buildup of these contaminant gases and water vapor and depletion of carbon dioxide during experiments. Most pollutant gases may be removed in water condensation on heat exchangers which set the dew point.

Chambers within growth rooms or within larger chambers also have the advantage of controlled programmed environments. With several chambers housed in a larger facility, additional treatment levels can be administered at the same time during an experiment. Chambers within large units may also permit greater flexibility in optimizing ventilation rates to meet plant needs. Replication both within and among runs is facilitated by this kind of equipment. Disadvantages include the limited size of chambers that can be accommodated. The artificial conditions created in all controlled environment chambers may be unsatisfactory for studies which require close simulation of outdoor conditions.

Continuous stirred tank reactor chambers (CSTR's) have distinct advantages for gas exchange studies because fluxes can be readily calculated. Chambers can be placed in greenhouses or growth rooms. The rapid air mixing minimizes gradients within chambers as well as leaf boundary layer resistance. In terms of disadvantage, the lack of gradients of temperature, humidity, and gaseous pollution may not represent natural ecosystem conditions. The rapid air movement may cause wind injury to sensitive plants. The limited size of these chambers, as constructed to date, will be a disadvantage in studies with large plants. Lighting systems are often a problem and appropriate minimum light levels and temperature- and humidity-controlled air will need to be supplied to each chamber.

5. Cuvettes

Four basic types of cuvettes are utilized in gas exchange systems:

Type 1: Cuvettes for Sampling Rates of Gas Absorption or Emission from Leaves;

Type 2: Cuvettes for Continually Monitoring the Amount of Gas Absorption or Emission from Leaves -- Differential Gas Exchange System;

Type 3: Cuvettes for Maintaining Constant Levels of Water Vapor and CO₂ -- Null Balance, Steady State Gas Exchange Systems;

Type 4: Cuvettes for Carbon Isotope Studies.

The basic descriptions of these systems are presented in Chapter II. In Table IV-5 we evaluate the features usually associated with each of these systems and classify some well-known models of cuvettes/gas exchange systems. This table can be used to evaluate the potential for several gas exchange systems to be used for a wide range of purposes including maintenance of environmental conditions which can affect foliar air pollution absorption characteristics. In addition, the gas exchange systems are evaluated for long and short-term experimental capability as well as the capability for controlled air pollution exposures. The evaluation of single leaf cuvette systems as fumigation devices has been limited because cuvettes are designed to have high degrees of gas mixing, to have uniform distribution around the leaf, and to have the smallest leaf boundary layer possible. Also, the small volume of these cuvettes (usually less than 1 liter) makes direct measurements of gradients in gas concentrations across a leaf surface a difficult technical task. Larger laboratory fumigation chambers which may contain one or several whole plants have complex patterns of atmospheric mixing which reflects air flow rates, chamber shape and size, physical surfaces in the chamber such as chamber walls, pots, soils, as well as the distribution of plants, plant architecture, and environmental conditions in the chamber (Unsworth and Mansfield, 1980). Thus, whole plant chambers must be thoroughly investigated in order to understand air pollution fluxes between the atmosphere, plants, and other surfaces; failure to do so may result in misleading expressions of air pollution exposures and other gas exchange characteristics.

B. Wet Deposition

The performance evaluation of exposure systems simulating rainfall, cloud water, and aerosols is presented in Table IV-6. Eight generic systems of rainfall simulation are evaluated, including indoor systems and four types of systems for outdoor exposures (no exclusion of ambient rainfall, permanent exclusion, automated exclusion/scheduled addition, and automated exclusion/automated addition of rain). The general performance of each type of system is characterized according to the criteria outlined in Section III. Because some of the criteria are not important for selected systems, the evaluation process provides two-fold remarks to indicate relevance and degree of characterization reported (relevant/characteristics).

Table IV-5. Performance evaluation for cuvette/gas exchange systems.

	1	Cuvette Type ^a			4
		2	3		
Evaluation of Operational Characteristics					
Closed air flow system	Y ^b	N	Y		Y
Flow-through air flow system	N	Y	Y		N
Temperature control	N	Y	Y		N
Humidity control	N	Y	Y		N
Clip-on cuvette	Y	N	Y		Y
Whole-leaf cuvette	Y	Y	Y		Y
Short-term measurements only	Y	N	N		Y
Long-term measurement capability	N	Y	Y		N
Diagnostic gas exchange capability	N	Y	Y		Y
Air pollution fumigation capability	N	Y	Y		N
Classification of Some Models					
Diffusive resistance porometer (numerous models)	X				
Analytical Developmental Corp. Delta T System	X				
LiCor steady state porometer (1600)				X	
LiCor photosynthesis system (6000 and 6200)	X				
Sieman cuvette systems		X			
Data Design Group system (PACsys 9900)				X	
Analytical Development Corporation system (LCA-2)		X			
Armstrong Enterprises system (custom design)		X	X		
Walz system (custom design)		X			

^a See text.

^b Important, Y = Yes, N = No for this type of cuvette.

Table IV-6. Performance evaluation for wet deposition systems.

	Rainfall Simulation							
	Outdoor Systems							Aerosol Simulation
	Indoor Systems	Shelterless Scheduled Addition	Permanent Shelter Scheduled Addition	Automated Exclusion Scheduled Addition	Automated Exclusion Automated Addition	Combination Wet and Dry	Cloud Water Simulation	
A. Hydrometeor/Pollutant: Chemistry and Physics								
1. Simulant Chemistry	Y/Y ^a	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y
2. Incident Hydrometeor Chemistry								
a. Spatial Features	Y/N	Y/N	Y/N	Y/N	Y/Y	Y/Y	Y/N	Y/N
b. Temporal Features	Y/Y	N/Y	Y/N	Y/N	Y/Y	Y/Y	Y/N	Y/Y
3. Incident Hydrometeor Deposition								
a. Spatial Features	Y/N	Y/N	Y/N	Y/N	Y/Y	Y/Y	Y/N	Y/Y
b. Temporal Features	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y
4. Chemical-Specific Deposition Rates								
a. Spatial Features	Y/N	N/N	N/N	N/N	N/N	N/N	Y/N	Y/Y
b. Temporal Features	Y/N	Y/N	Y/N	Y/N	Y/Y	Y/Y	Y/N	N/N
5. Incident Hydrometeor Physics	N/Y	Y/N	Y/N	Y/N	Y/Y	Y/Y	Y/N	N/N
6. Chemical Processing of Deposited Pollutants								
a. Foliar Retention	N/N	N/N	N/N	Y/N	Y/N	Y/Y	Y/Y	Y/Y
b. Throughfall/Stemflow	Y/N	Y/N	Y/N	Y/N	Y/N	Y/N	Y/Y	N/N
c. Soil	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	N/N
7. Pollutant Mixtures (Wet/Wet and Wet/Dry)	NA	NA	NA	NA	Y/Y	Y/Y	Y/Y	NA
B. The Environment: Physical, Chemical, and Biological Features								
1. Atmosphere								
a. Radiation	N/N	N/N	Y/Y	Y/N	Y/N	Y/N	N/N	Y/Y
b. Temperature/Moisture	N/N	Y/N	Y/N	Y/Y	Y/N	Y/N	N/Y	Y/Y
c. Turbulence/Mixing	N/N	N/N	Y/Y	Y/Y	Y/Y	Y/Y	N/N	Y/Y
d. Trace Gases (Pollutant and Nonpollutant)	Y/N	Y/N	Y/Y	Y/Y	Y/Y	Y/Y	N/N	Y/N
2. Vegetation								
a. Structure	N/N	N/N	N/N	N/N	N/N	N/N	Y/Y	Y/Y
b. Physiology and Growth	N/N	Y/N	Y/N	Y/Y	Y/Y	Y/Y	N/N	Y/Y
3. Soil	Y/Y	Y/Y	N/N	Y/Y	Y/Y	Y/Y	N/N	N/N
D. Hardware								
1. Hydrometeor Dispens.	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y
a. Droplet Distrib.	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y
b. Deposition Mont. and Control	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y
2. Environment								
a. Monitoring	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y	Y/Y
b. Control	N/N	N/N	Y/N	Y/N	Y/N	Y/N	Y/N	Y/Y
3. Data Acquisition	N/N	N/N	N/N	N/N	Y/Y	Y/Y	N/N	N/N

^a Important/Characterized, Y = Yes, N = No for this type of system.

1. Indoor Rainfall Simulation -- Controlled-Environment or Greenhouse

The indoor exposure systems for rainfall are described in Appendix F, Section A. The principal advantage of rainfall simulation in a controlled environment is that by rigidly controlling exogenous physical, chemical, and biological factors, one can identify the role of individual factors (or combinations of factors) in governing the influence of wet deposition on the growth and physiology of wood plants. Consequently, these systems have superior capabilities for resolving mechanisms and processes governing plant response. The disadvantage of rigidly controlled exposures is the artificial nature of the exposure environment. In many agricultural and forested landscapes, the interaction of edaphic, climatic, and atmospheric factors during and after wet deposition events may play a substantial role in injury expression. Consequently, data and patterns observed in controlled exposures cannot be indiscriminately applied to predict effects of wet deposition in forests or agricultural settings.

2. Outdoor Rainfall Simulation -- Shelterless Plots

Shelterless plot systems experience simulated rain in excess of ambient levels and are described in Appendix F, Section B.1. The most significant advantage is their ability to accommodate variable plot sizes in terms of both surface area and canopy height. The absence of exclusion lids and the availability of relatively inexpensive irrigation systems makes this design worthy of consideration for many large scale projects. The use of these exposure systems in more pristine ecosystems has particular merit, especially in efforts to evaluate the effect of a particular chemical addition. The disadvantages include: (1) a requirement for a significant water conditioning and simulation mixing system (for large plot simulation); (2) difficulty in achieving uniform distribution, particularly in areas experiencing moderate winds; and (3) the interpretational problem of evaluating the significance of excess water and chemical input to plant growth. This interpretation should not be regarded as simply an additive factor, for it may exhibit characteristics of joint action (i.e., synergism, antagonism).

3. Outdoor Rainfall Simulation -- Permanent Sheltered Plots

Sheltered plot exposure systems are described in Appendix F, Section B.2. Unlike controlled-environments, sheltered plots under field conditions offer many features of the environment at near ambient levels. Consequently, these systems can be used to test general hypotheses on the influence of wet deposition on woody vegetation. The resulting data can help establish priorities for subsequent research by identifying hypotheses worthy of more careful evaluation under more realistic field conditions. The major disadvantage is that some critical environmental factors cannot be rigidly controlled, including temperature, light, and moisture conditions. In all systems, rainfall simulation is by scheduled addition.

4. Outdoor Rainfall Simulation -- Automated Exclusion/Scheduled Addition of Rainfall

The automated exclusion systems are described in Appendix F, Section B.3. The major advantage of these systems is their ability to maintain a near-normal microclimate (edaphic plus atmospheric) during non-exposure periods, since the exclusion lids are positioned away from the canopy. Consequently, the systems provide more realistic data with which to evaluate the influence of wet deposition on plant growth and development. The major disadvantage is the prescheduled protocol for rainfall simulation so that simulated rain events may or may not co-occur with ambient. Because the canopy's microclimate experiences wet deposition on a more frequent basis than ambient plots (e.g., saturated vapor pressure, reduced temperature, light attenuation), these systems may present interpretational problems in evaluating the dose-response functions. A second, less important disadvantage is the stability of the exclusion track system in high winds.

5. Outdoor Rainfall Simulation -- Automated Exclusion/Automated Addition of Rainfall

The chief advantage of these systems described in Appendix F, Section B.4, is that the canopy's microclimate experiences wet deposition events that mimic ambient conditions including both the time of delivery and the deposition rate. The simultaneous exclusion and addition is a significant advantage. The disadvantages are: (1) the system may inadvertently modify the microclimatic conditions through other hardware devices (e.g., open-top chamber); (2) the fabrication and maintenance costs are significant; and (3) some experimental designs cannot be accommodated with automated addition given the natural variation in rainfall.

6. Facilities for Simulating Wet and Dry Deposition

These systems combine various individual methods of wet and dry simulation to achieve the objective. The systems available for this type of mixture exposure studies are described in Appendix D. They have the advantage of being able to partition sources of phytotoxicity to either wet or dry deposition, singly and in combination. The chief disadvantage is the additional hardware required to maintain the system throughout the growing season, and the inadvertent modification of the exposure environment which comes with the exclusion of ambient pollutant.

7. Cloud Water/Mist Simulation

Most cloud water system designs are primitive (Appendix G) and unsuitable for field application, particularly in areas with advective cloud water. Existing laboratory facilities are capable of evaluating potential mechanisms of phytotoxicity. The resulting data, however, should not be indiscriminately applied to predict effects under ambient conditions.

8. Aerosol Simulation

The significance of aerosols in terms of vegetation response remains a low priority. The most important issues are likely to be deposition and fate of particular chemicals in the aerosol. The disadvantage of these systems is the need for a rigidly controlled growth environment, commonly provided through a dedicated chamber. The available systems are described in Appendix G.

V. Exposure Protocol/Air Quality

A discussion of exposure protocols for deposition studies involves first the acceptance that exposure studies must relate, at least at some level, to the characteristics of air quality in the region, site, or elevation of interest for the particular forest species under study. The characteristics of the ambient air quality can be used to develop the appropriate exposure regime(s)/protocol(s), whether for exposure-response studies in the field or for controlled-exposure studies.

Reliable, up-to-date monitoring programs and accessible data bases for both wet and dry deposition are crucial to understanding the nature of occurrences of the pollutant(s) and for developing exposure protocols. Toward this end, the availability and completeness of available monitoring data are reviewed with details of the databases included in Appendix J.

Characterization of air quality to establish the dynamic elements of biological exposures, including temporal and spatial distributions of the pollutant(s) concentrations, has only recently been the subject of attention for biological studies, and thus there is not an extensive literature base. In contrast, annual trends and compositional information for both wet and dry deposition have been extensively reviewed (e.g., USEPA, 1984; USEPA, 1982; Altshuller, 1984). Highlights of these general trends of air quality are included in Appendix K. In this chapter only the general temporal and spatial distribution characteristics observed for the various pollutants concerned with forest health are discussed.

The types of exposure regimes used historically in both dry and wet deposition studies are discussed, including the air quality characterization information needed in their development, the degree to which the exposure protocols relate to current air quality, and the limitations in interpretation of the experimental results imposed by their use.

A. Data Bases -- Dry (Gaseous) and Wet Deposition

Over the years, a number of programs have measured ambient air quality and wet deposition in various areas of the United States. The sampling has ranged from short-term (a few months) to multi-year projects. The sampling programs have had disparate goals, consequently, the monitoring data do not have the same level of resolution, are not readily available, and have not all been entered into data bases. Another consequence of the disparate objectives is that monitoring has been done extensively in some areas of the country with little or no data available for other locations.

Gaseous Air Pollutants: Dry deposition is the direct transfer of compounds (gases or particles) from the atmosphere to vegetation or soil surfaces. Gaseous air pollutants have been more extensively monitored than the dry particle deposition. The data are usually reported as hourly mean concentrations. Most of the available monitoring data for gaseous air pollutants are from sites in urban or near-urban areas or in the vicinity of major pollution emitters. There are several readily available sources of air quality monitoring databases such as; EPA-SAROAD, EPRI, TVA, and the National Park Service

that can be used to obtain information concerning potential vegetation exposures. The data bases are discussed in detail in Appendix J. Individual investigators may be able to locate other site-specific data from various agencies or research projects that will assist in their studies.

B. Characteristics of Ambient Air Quality in the U.S.

The hypotheses that invoke various dry and wet deposition components, individually or in combinations, to explain the observed decline in forest species requires first demonstrating the occurrence and general trend of these various pollutants in the regions, sites, and/or elevations of interest. Testing these hypotheses further requires an understanding of the dynamics of the various components and characteristics of ambient air quality, including the temporal and spatial distribution of concentrations over specific time periods, for developing experimental exposure regimes. Chronic exposure studies, specifically, need to incorporate the elements of temporal dynamics of the pollutant(s) in question into the experimental protocol.

Although reasonably good monitoring databases are available for characterization of current air quality over most of the regions of the United States, the long-term trends in dry and, especially wet deposition, in the U.S. are difficult to assess because of the lack of a really significant historical monitoring database for any of the various pollutants. Given this limitation, general trends of the various component pollutants in both wet and dry deposition have been reported and are briefly revised in Appendix K. These trends in pollutant concentrations are important as a first level understanding of the dynamics of air quality; and the incorporation of this information into experimental protocols for exposure studies of forest species requires some means of characterization of the air quality data that is relevant to the postulated effect on tree vigor.

Air Quality Characterization: Dry Deposition

Characterization of air quality with long-term mean concentrations such as annual, quarterly, monthly, and even 7-hr means is the most prevalent approach, but has questionable biological relevance. An average of hourly concentrations over a set time period minimizes the contributions of the peak concentration values, which is inconsistent with the literature describing the importance of peaks in the plant response to a pollutant such as ozone (e.g., Heck et al., 1966; Amiro et al., 1984). Other studies have shown the importance of the spatial and temporal distributions of the hourly concentrations of ozone in plant growth (e.g., Musselman et al., 1983; Hogsett et al., 1985). The relationship of air quality to the biological response of plants requires more detailed analyses of ambient air to understand the spatial and temporal distributions of concentrations of the pollutant(s) in question. Characterization to define these temporal patterns and concentration-frequency occurrences provides not only the starting point for relating deposition exposure studies and the actual air quality of interest across the various regions of the U.S., but also the information needed to design experiments that reflect the temporal and spatial nature of the region, site or elevation of interest (Lefohn et al., 1986a,b). A particular site for one particular year can be characterized; another approach is to characterize a site over a number of years or a number

of sites over a region by calculating average characterization information with standard deviations giving variation over years or regions. The hourly monitoring data is characterized with the following criteria: (1) percentile distribution of hourly concentrations; (2) number of hourly occurrences equal to or greater than a specified minimum concentration; (3) duration of the episodes or pre-defined peak concentrations; (4) respite time between these episodes; (5) average diurnal pattern (over the growing season or annually) of hourly concentrations for daily temporal distribution of concentrations and identifying the time of day when the daily maximum occurs. Such characterization would describe, in large part, the chemical and/or temporal pollutant under study.

Ozone

Ozone percentile distributions from a number of sites across the country are shown in Table V-1. Most of the sites, with the exception of the California sites, are similar in distributions at the 95th percentile. In addition, the year-to-year variation at a given site is small, varying only 10-15% at the 95th percentile and even less at the 50th percentile. All sites display an infrequent occurrence of the higher concentration values; 70% of the time, hourly concentrations of 0.050 ppm or less are reported. The relative infrequency of the higher concentrations is also shown in histograms of the number of hourly occurrences at specified minimum values (Figure V-1). Rural sites in the upper Midwest and the Northeast are similar (Figure V-1a, 1b), as well as sites in the Southeast which are not shown. Hourly concentrations of 0-0.03 ppm occur most frequently and values greater than 0.08 ppm occur infrequently over a growing season. A remote site in the Shenandoah National Park at 3500 ft. has a somewhat different distribution (Figure V-1c); the majority of hourly occurrences are around 0.05-0.06 ppm, rather than the log-normal shape of the rural site distributions under influence of anthropogenic sources. These differences are not necessarily a function of remote versus rural site designation, or a function of elevation, but simply an observation of two different types of concentration distributions. These distributions are also shown in diurnal plots in Figures V-2 and V-3. The spatial pattern of occurrence of these higher concentrations or episodes is important in biological response and requires a knowledge of the duration of the episodes, the probability of the episode occurrence, and the length of time between the episodes or respite times. Analysis of rural and remote sites in the Midwest, Great Lakes states, Northeast, and Southeast (Lefohn *et al.*, 1986a; 1986b) indicates that all regions display average growing season patterns of hourly concentrations where occurrences of higher concentrations are infrequent; the episodes or peaks lasting relatively short periods and time between these episodes being relatively long. Duration of episodes and the time between episodes or respite times for a number of sites in the Northeast and Southeast are given in Tables V-2 and V-3, respectively. Using a different approach in analysis of air quality, Taylor and Norby (1985) calculated the probability of episodes and periods of respite lasting defined time periods (days). The analysis was based on a 4-year monitoring data set from the Shenandoah collected by Skelly and co-workers. In their analysis, an episode was defined as any day(s) in which a 1-hr ozone concentration exceeded 0.08 ppm. The Shenandoah National Park monitoring site experienced an average of five episodes over the growing season for the 4-year monitoring period. Single-day episodes were infrequent ($P = 16\%$), but there was an 80% probability

Table V-1. Ozone percentile distribution in some rural sites across the U.S. in the SAROAD data base (concentration in ppm). From Lefohn and Benedict (1985); Lefohn (personal communication).

Site	State	Year	Min. Obs.	Percentile						Max. Obs.
				10	30	50	70	90	95	
Fontana (RC) ^a	CA	1980	0.003	0.010	0.020	0.030	0.050	0.110	0.160	0.420
Fontana (RC)	CA	1981	0.003	0.003	0.010	0.020	0.040	0.010	0.160	0.350
Fresno Co. (RE)	CA	1979	0.003	0.030	0.050	0.060	0.070	0.090	0.090	0.150
Oxnard (RA)	CA	1979	0.003	0.003	0.010	0.020	0.040	0.060	0.070	0.230
Riverside (RA)	CA	1981	0.003	0.003	0.010	0.020	0.040	0.100	0.130	0.280
San Bernardino Co. (RNU)	CA	1979	0.003	0.010	0.030	0.040	0.070	0.140	0.170	0.400
San Bernardino Co. (RNU)	CA	1981	0.003	0.010	0.020	0.040	0.060	0.120	0.150	0.350
Ventura Co. (RA)	CA	1979	0.003	0.003	0.020	0.030	0.040	0.080	0.100	0.220
Columbia Co. (WI)	WI	1979	0.002	0.018	0.034	0.044	0.054	0.072	0.085	0.172
Juneau (WI)	WI	1979	0.002	0.006	0.024	0.035	0.045	0.060	0.070	0.125
Sherburne Co. (MN)	MN	1980	0.000	0.010	0.022	0.030	0.038	0.050	0.058	0.115
Will Co. (IL)	IL	1981	0.000	0.006	0.018	0.029	0.039	0.055	0.063	0.118
Allen Co. (IN)	IN	1981	0.005	0.005	0.013	0.023	0.035	0.054	0.066	0.108
Porter Co. (IN)	IN	1982	0.003	0.003	0.018	0.030	0.040	0.060	0.068	0.142
Kent Co. (MI)	MI	1981	0.000	0.013	0.024	0.030	0.040	0.057	0.066	0.108
Preble Co. (OH)	OH	1981	0.000	0.005	0.017	0.030	0.040	0.060	0.072	0.127
Hamilton Co. (RI)	OH	1978	0.003	0.003	0.003	0.010	0.025	0.055	0.077	0.180
Hamilton Co. (RI)	OH	1980	0.003	0.003	0.003	0.012	0.025	0.055	0.072	0.172
Madison Co. (RNU)	IL	1978	0.005	0.005	0.014	0.027	0.041	0.069	0.083	0.187
Madison Co. (RNU)	IL	1980	0.003	0.003	0.014	0.025	0.035	0.062	0.077	0.170
Jefferson Co. (RNU)	KY	1978	0.003	0.003	0.016	0.027	0.042	0.064	0.076	0.182
Jefferson Co. (RNU)	KY	1980	0.003	0.003	0.016	0.030	0.047	0.077	0.093	0.186
Clay Co. (RNU)	MO	1980	0.003	0.010	0.020	0.030	0.041	0.060	0.070	0.160
Omaha (RNU)	NE	1980	0.003	0.010	0.020	0.030	0.042	0.070	0.085	0.170
Mobile (R)	AL	1978	0.000	0.000	0.007	0.022	0.040	0.060	0.070	0.120
Hillsborough Co. (R)	FL	1978	0.000	0.008	0.015	0.025	0.035	0.052	0.062	0.138
St. Petersburg (R)	FL	1979	0.000	0.005	0.015	0.025	0.040	0.060	0.070	0.120
Pensacola (R)	FL	1981	0.000	0.001	0.015	0.027	0.040	0.056	0.064	0.112
Tarpon Springs (R)	FL	1981	0.000	0.005	0.015	0.030	0.040	0.060	0.070	0.115
Jefferson Co. (RNU)	AL	1981	0.003	0.003	0.007	0.030	0.049	0.074	0.086	0.166
North Little Rock (RNU)	AR	1979	0.003	0.012	0.024	0.034	0.046	0.067	0.081	0.144
Iberville Parish (RNU)	LA	1978	0.003	0.003	0.007	0.017	0.031	0.061	0.078	0.205
Arlington (RA)	TX	1979	0.003	0.010	0.030	0.040	0.050	0.070	0.080	0.190
Arlington (RA)	TX	1980	0.003	0.003	0.020	0.030	0.040	0.060	0.080	0.160
Denton Co. (RA)	TX	1981	0.003	0.003	0.010	0.030	0.040	0.060	0.080	0.150
Camden Co. (RA)	NJ	1979	0.003	0.003	0.015	0.025	0.035	0.060	0.073	0.147

(continued)

Table V-1 (continued)

Site	State	Year	Min. Obs.	Percentile						Max. Obs.
				10	30	50	70	90	95	
Camden Co. (RA)	NJ	1980	0.003	0.005	0.017	0.026	0.037	0.066	0.080	0.141
Anne Arundel Co. (R)	MD	1981	0.003	0.003	0.007	0.020	0.034	0.061	0.076	0.170
Greenwich (RNU)	CT	1979	0.003	0.005	0.013	0.024	0.034	0.054	0.070	0.204
Greenwich (RNU)	CT	1980	0.003	0.005	0.019	0.029	0.040	0.062	0.082	0.216
Agawam (RNU)	MA	1979	0.003	0.003	0.012	0.025	0.040	0.065	0.082	0.175
Worcester (SR)	MA	1979	0.000	0.019	0.028	0.036	0.046	0.064	0.080	0.185
		1981	0.000	0.021	0.032	0.040	0.050	0.067	0.078	0.139
Glen Falls (SR)	NY	1980	0.000	0.004	0.015	0.025	0.035	0.054	0.065	0.106
		1982	0.001	0.005	0.017	0.028	0.038	0.056	0.067	0.116
Kutztown (R)	PA	1982	0.000	0.003	0.017	0.030	0.046	0.071	0.085	0.150
		1983	0.000	0.003	0.019	0.032	0.045	0.071	0.086	0.134
Keene (CC)	NH	1982	0.000	0.005	0.018	0.029	0.041	0.059	0.069	0.135
Green Mountain (RNU)	VT	1980	0.000	0.005	0.020	0.035	0.050	0.065	0.075	0.115
		1981	0.000	0.005	0.015	0.030	0.040	0.055	0.060	0.105
Chequamegon N.F.	WI	1981	0.005	0.020	0.030	0.035	0.040	0.055	0.060	0.080
Croatan N.F. (RE)	NC	1980	0.000	0.010	0.020	0.030	0.045	0.065	0.075	0.150
		1982	0.000	0.005	0.020	0.025	0.040	0.055	0.060	0.095
Big Meadows Shen. N.P. (VA)		1983	0.005	0.030	0.045	0.055	0.065	0.075	0.080	0.010
Dickey Ridge Shen. N.P. (VA)		1983	0.000	0.030	0.045	0.055	0.065	0.075	0.085	0.110
Sawhill Run Shen. N.P. (VA)		1983	0.000	0.020	0.030	0.040	0.055	0.070	0.080	0.120
Mark Twain N.F. (MO)		1981	0.000	0.015	0.025	0.030	0.040	0.050	0.060	0.115
Mark Twain N.F. (MO)		1983	0.000	0.025	0.035	0.045	0.055	0.070	0.080	0.110

RE = Remote; RNU = Rural Near Urban; RA = Rural Agricultural; RC = Rural Commercial;
 RI = Rural Industrial; R = Rural (unspecified).

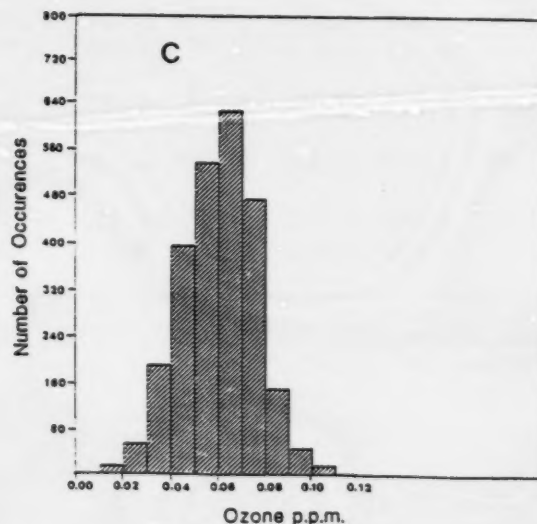
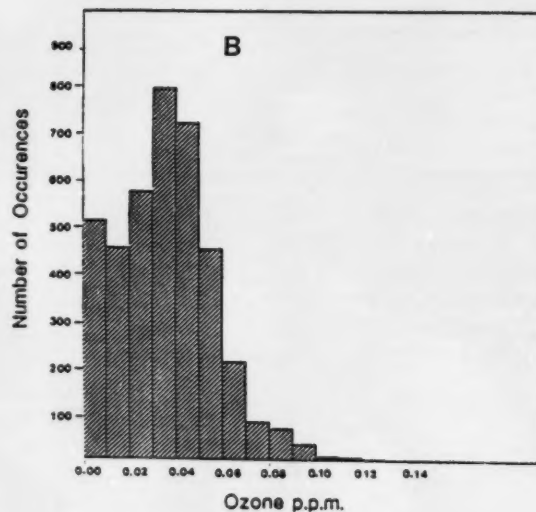
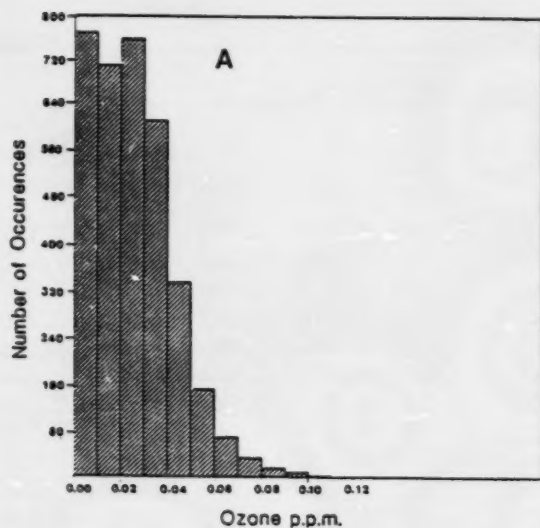


Figure V-1. Histograms of number of hourly occurrences of ozone at specified concentrations taken from monitoring data of rural and remote sites. A. Glen Falls, New York (May-September 1983); B. Juneau, Wisconsin (April-October 1979); C. Big Meadows, Shenandoah National Park, Virginia (May-September 1983). From Lefohn (personal communication).

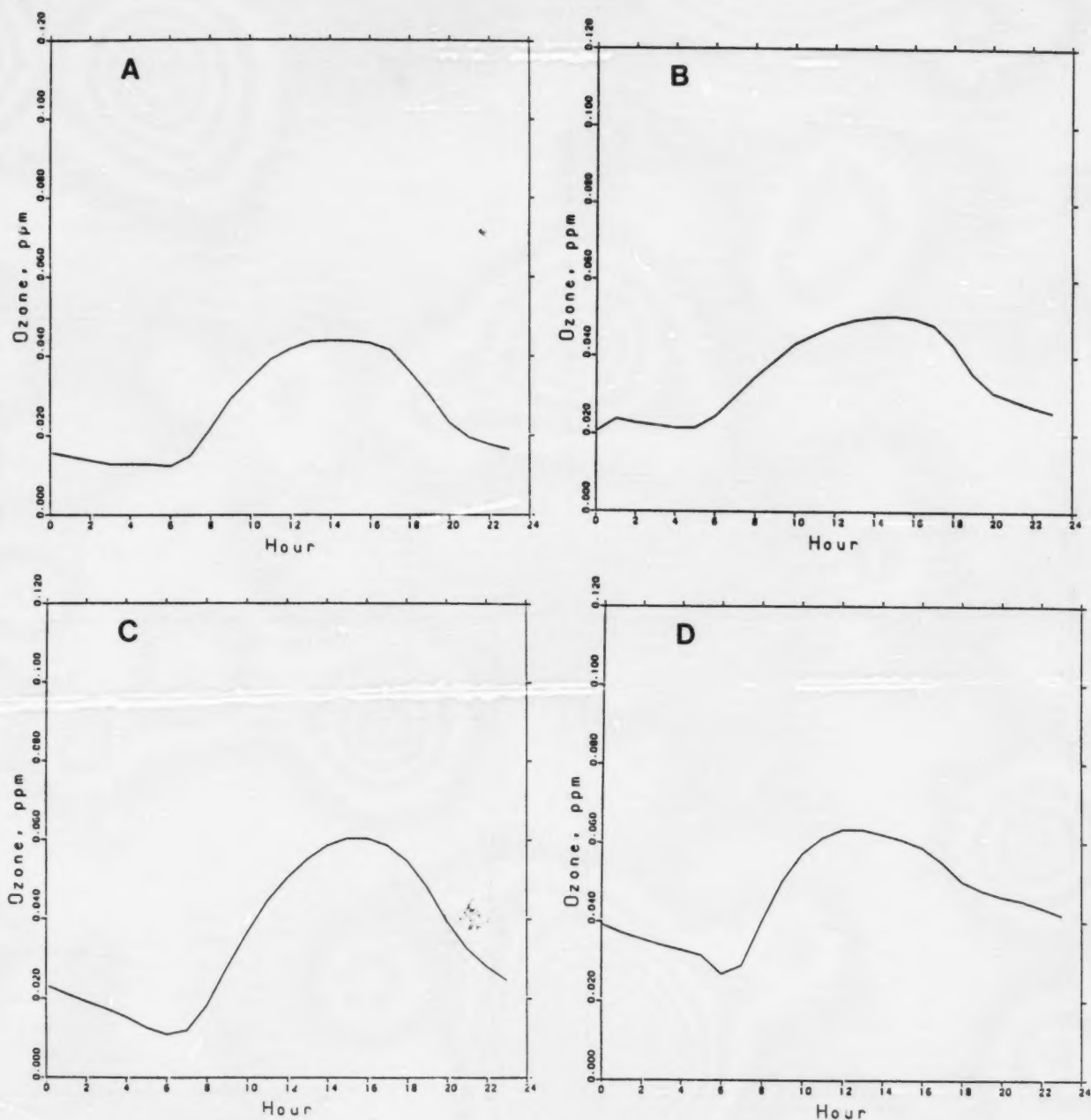


Figure V-2. Average diurnal concentration pattern from rural and remote sites. A. Croatoan National Forest, North Carolina (May-September 1983); B. Juneau, Wisconsin (April-October 1979); C. Kutztown, Pennsylvania (May-September 1983); D. Sawmill Run, Shenandoah National Park (May-September 1983). From Lefohn (personal communication).

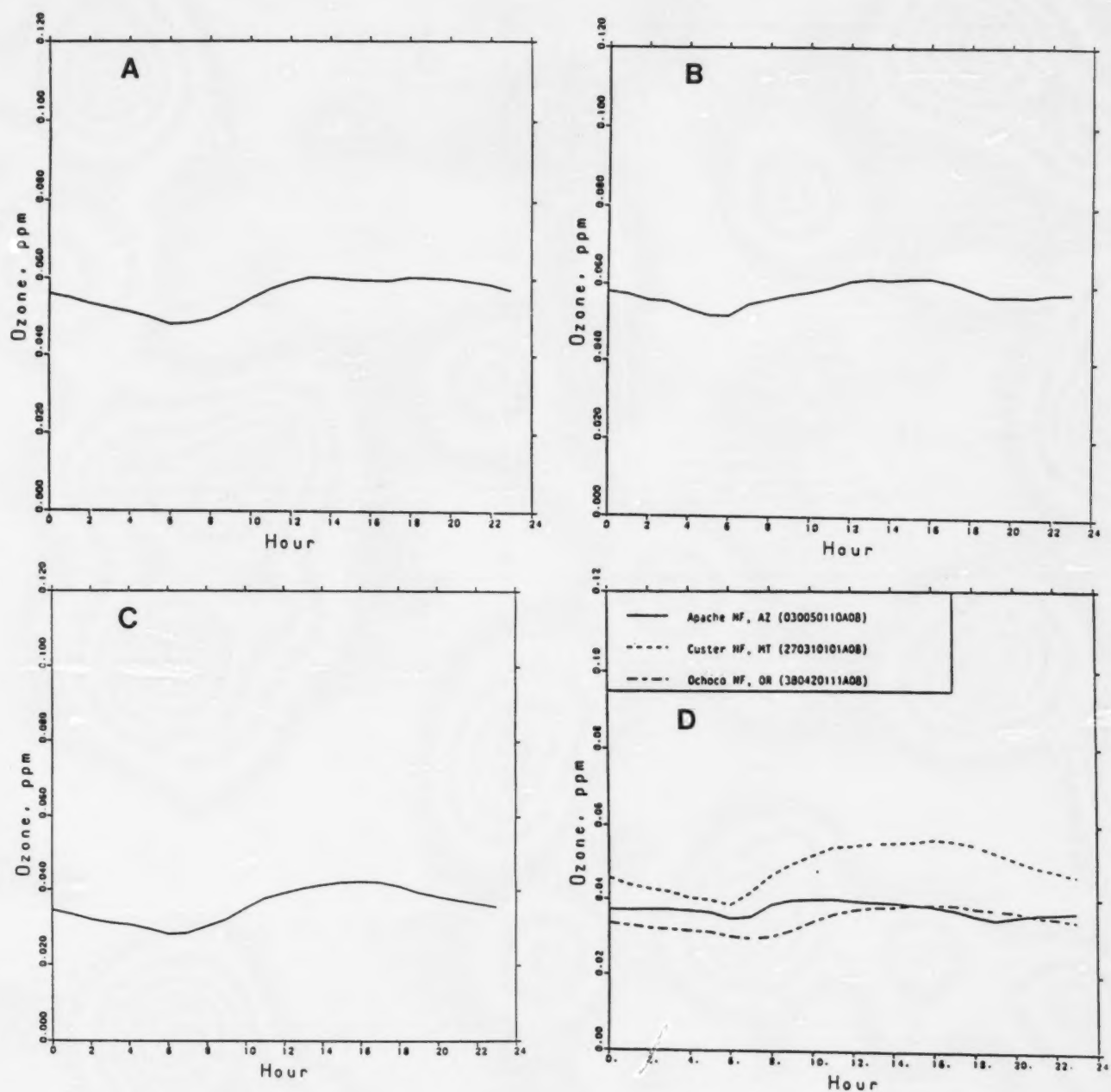


Figure V-3. Average diurnal concentration patterns from remote monitoring sites. A. Dickey Ridge, Shenandoah National Park, Virginia (May-September 1983); B. Big Meadows, Shenandoah National Park (May-September 1983); C. Chequamegon National Forest, Wisconsin (April-October 1981); D. Three National Forest sites (June-August 1981).

Table V-2. Average duration (hrs) of events that meet or exceed the indicated concentration, and the number of events (in parentheses) (Lefohn, personal communication).

	Average Length of Events That Meet or Exceed the Ozone Concentration			
	ppm			
	0.05	0.07	0.09	0.12
Worcester (1979)	7.9 (113)	5.9 (47)	3.3 (29)	2.0 (5)
Glen Falls, NY (1980)	4.8 (87)	3.4 (36)	2.9 (11)	0 (0)
Kutztown, PA (1983)	67 (131)	5.8 (63)	3.7 (35)	1.6 (8)
Keene, NH (1982)	6.4 (107)	5.1 (30)	3.2 (13)	2.0 (1)
Green Mountain N.F., VT (1981)	6.8 (61)	4.4 (10)	3.5 (2)	0 (0)
Croaton N.F., NC (1983)	6.1 (124)	3.3 (18)	0 (0)	0 (0)
Dickey Ridge, Shenandoah National Park (1983)	19.9 (95)	8.2 (114)	4.3 (31)	0 (0)
Big Meadows, Shenandoah National Park (1983)	19.6 (95)	6.2 (111)	3.1 (20)	0 (0)
Sawmill Ridge, Shenandoah National Park (1983)	8.8 (162)	5.3 (84)	3.4 (26)	2.0 (1)

Table V-3. Average respite time (hr) between episodes which equal or exceed indicated ozone concentration (Lefohn, personal communication).

	ppm			
	0.05	0.07	0.09	0.12
Worcester, MA (1979)	22.9	63.9	110.8	789.5
Glen Falls, NY (1980)	33.2	89.6	251.0	---
Kutztown, PA (1983)	19.6	44.1	68.2	290.1
Keene, NH (1982)	27.3	106.7	254.8	---
Green Mountain N.F., VT (1981)	47.4	219.7	---	---
Croatan N.F., NC (1983)	46.2	194.4	---	---
Dickey Ridge, Shenandoah National Park (1983)	9.4	22.2	92.3	---
Big Meadows, Shenandoah National Park (1983)	17.8	25.1	130.6	---
Sawmill Run, Shenandoah National Park (1983)	11.3	27.6	83.4	---

that the episode would last 2 or more days. A respite time of less than 2 weeks had a probability of 50%, and the median respite time between episodes was 10 days. The seasonality of peak occurrences was also suggested by this analytical approach. An episode was most likely to occur during May, June, and July (about equally), 40% less likely to occur during August and even less likely during the fall months. In general, the episodes occur infrequently and with less duration than the respite times (< 0.08 ppm) between the episodes.

An additional temporal and spatial feature important in analysis of air quality and the response of vegetation is the diurnal concentration pattern. How these hourly concentration values are distributed over a day is shown in average diurnal plots (Figures V-8 and V-9). The majority of sites analyzed display a diurnal rise and fall in concentration with the maximum occurring in the afternoon hours, obviously closely tied to diurnal climatic patterns and influenced by nearby anthropogenic sources (Figure V-8). Another diurnal pattern observed at some remote sites is one in which there is very little, if any, rise and fall in concentration; and most hours of the day experience concentrations above 0.03 ppm (Figure V-3). With these sites no clearly discernible maximum concentration is observed. The percentile distributions for these sites show 80-90% of the hourly values equal to or above 0.030 ppm, but still relatively infrequent occurrence of values above 0.080 ppm over the growing season (Table V-1). This lack of diurnal variation in concentration has also been associated with increased elevation in the U.S. and Germany (Taylor and Norby., 1985; Blank, 1985) and attributed to the reduction in atmospheric scavenging processes at higher elevation (e.g., nitric oxides). The explanation for this elevational distribution pattern is not clear and caution is urged in generalizing this observed phenomenon until further monitoring data is available from a variety of sites and elevational gradients. The diurnal patterns shown in Figures V-2 and V-3 are from monitoring sites located at elevations of 13 m (Croatan N.F., N.C.), 440 m (Chequamegon, N.F., MI), 1200 m (Custer N.F.); 1500 m (Sawmill Run S.N.P.), 2100 m (Dickey Ridge, S.N.P.), 2500 (Apache N.F.), and 3500 m (Big Meadows, S.N.P.). With these examples, the lack of diurnal variation does not hold true for all increased elevation sites.

Sulfur Dioxide

In the case of SO_2 , the mean values describing regional air quality are most often derived from point-source monitoring sites, and it is difficult to estimate how relevant these data are for quantifying air quality in regional terms. Point-source SO_2 concentration and frequency of occurrence depend upon the emission strength, meteorological conditions (e.g., wind speed), and distance between the source and monitoring site, thus bringing into question the use of these data to characterize of air quality in assessing SO_2 impact on forests located 25-50 miles from a source. There remains a need for site-specific monitoring to characterize SO_2 impact.

The percentile distribution from a number of SO_2 monitoring sites indicates 70% or more of the hourly values are below 0.01 ppm (Table V-4). This distribution points out the ineffectiveness of an annual mean for characterizing SO_2 . An example of the distribution of hourly concentration values for SO_2 is given in Figure V-4 and, unlike the lognormal-like distribution for ozone at

Table V-4. Percentile distribution for sulfur dioxide (ppm), January-December. From Lefohn et al. (1986b).

Site	State	Year	Min. Obs.	Percentile							Max. Obs.
				10	30	50	70	90	95	99	
North Little Rock (RNU) ^a	AR	1981	0.000	0.000	0.000	0.000	0.000	0.005	0.007	0.015	0.100
North Little Rock (RNU)	AR	1982	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.008	0.021
Pensacola (RNU)	FL	1981	0.000	0.000	0.001	0.003	0.006	0.014	0.028	0.107	0.415
Pensacola (RNU)	FL	1982	0.000	0.000	0.002	0.005	0.008	0.018	0.027	0.095	0.280
Tarpon Springs (RC)	FL	1981	0.000	0.000	0.000	0.000	0.000	0.010	0.015	0.040	0.145
Tarpon Springs (RC)	FL	1982	0.000	0.000	0.000	0.000	0.000	0.005	0.010	0.035	0.145
Trigg Co.	KY	1982	0.001	0.001	0.001	0.002	0.004	0.011	0.017	0.032	0.103
Trigg Co.	KY	1983	0.000	0.001	0.001	0.002	0.004	0.011	0.016	0.031	0.105
Jay (R)	ME	1982	0.001	0.002	0.002	0.003	0.004	0.008	0.011	0.022	0.194
Jay (R)	ME	1983	0.002	0.002	0.002	0.003	0.004	0.006	0.008	0.020	0.085
Rumford (005JOS) (RI)	ME	1983	0.000	0.001	0.002	0.008	0.013	0.036	0.059	0.126	0.313
Pittsfield (RI)	MA	1982	0.000	0.000	0.002	0.005	0.010	0.018	0.025	0.055	0.120
Lake Co.	MN	1978	0.000	0.002	0.002	0.002	0.002	0.002	0.002	0.002	0.030
Lake Co.	MN	1979	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Iron Co. (RI)	MO	1982	0.000	0.000	0.000	0.000	0.006	0.020	0.030	0.117	1.000
Iron Co. (RI)	MO	1983	0.000	0.000	0.000	0.003	0.007	0.019	0.034	0.120	0.565
Berlin (RI)	NH	1983	0.000	0.000	0.000	0.002	0.005	0.016	0.023	0.051	0.120
Pembroke (RNU)	NH	1983	0.000	0.000	0.000	0.000	0.004	0.013	0.031	0.092	0.294
Camden Co.	NJ	1982	0.000	0.002	0.003	0.004	0.009	0.016	0.020	0.036	0.088
Essex Co. (RE)	NY	1983	0.000	0.000	0.002	0.003	0.004	0.006	0.007	0.014	0.044
Rensselaer (SI)	NY	1980	0.000	0.001	0.004	0.009	0.015	0.029	0.040	0.067	0.228
Rensselaer (SI)	NY	1982	0.000	0.001	0.004	0.007	0.012	0.026	0.036	0.059	0.144
Columbus Co. (RI)	NC	1982	0.003	0.003	0.003	0.003	0.003	0.076	0.015	0.039	0.141
Columbus Co. (RI)	NC	1983	0.003	0.003	0.003	0.003	0.003	0.009	0.014	0.029	0.086
Monroe Co. (RE)	TN	1979	0.000	0.000	0.000	0.000	0.000	0.010	0.010	0.030	0.080
Monroe Co. (RE)	TN	1980	0.000	0.000	0.000	0.000	0.010	0.010	0.020	0.030	0.060
Burlington (CC)	VT	1982	0.001	0.001	0.002	0.005	0.007	0.015	0.020	0.032	0.165
Dickey Ridge, Shen. (NP)	VA	1983	0.000	0.000	0.000	0.006	0.005	0.010	0.015	0.025	0.060
Salem	VA	1982	0.000	0.002	0.003	0.006	0.005	0.010	0.015	0.025	0.065
Lewisburg	WV	1979	0.000	0.000	0.000	0.000	0.003	0.011	0.017	0.037	0.112
Chippewa Co. (RA)	WI	1980	0.001	0.001	0.001	0.001	0.001	0.001	0.002	0.004	0.021
Porter Co. (R)	IN	1982	0.001	0.001	0.001	0.003	0.007	0.014	0.022	0.046	0.184

^a All sites are 0-60 miles from a National Forest. RE = remote; RNU = rural near urban; RA = rural agricultural; RC = rural commercial; RI = rural industrial; R = rural unspecified; CC = city center; NP = National Park.

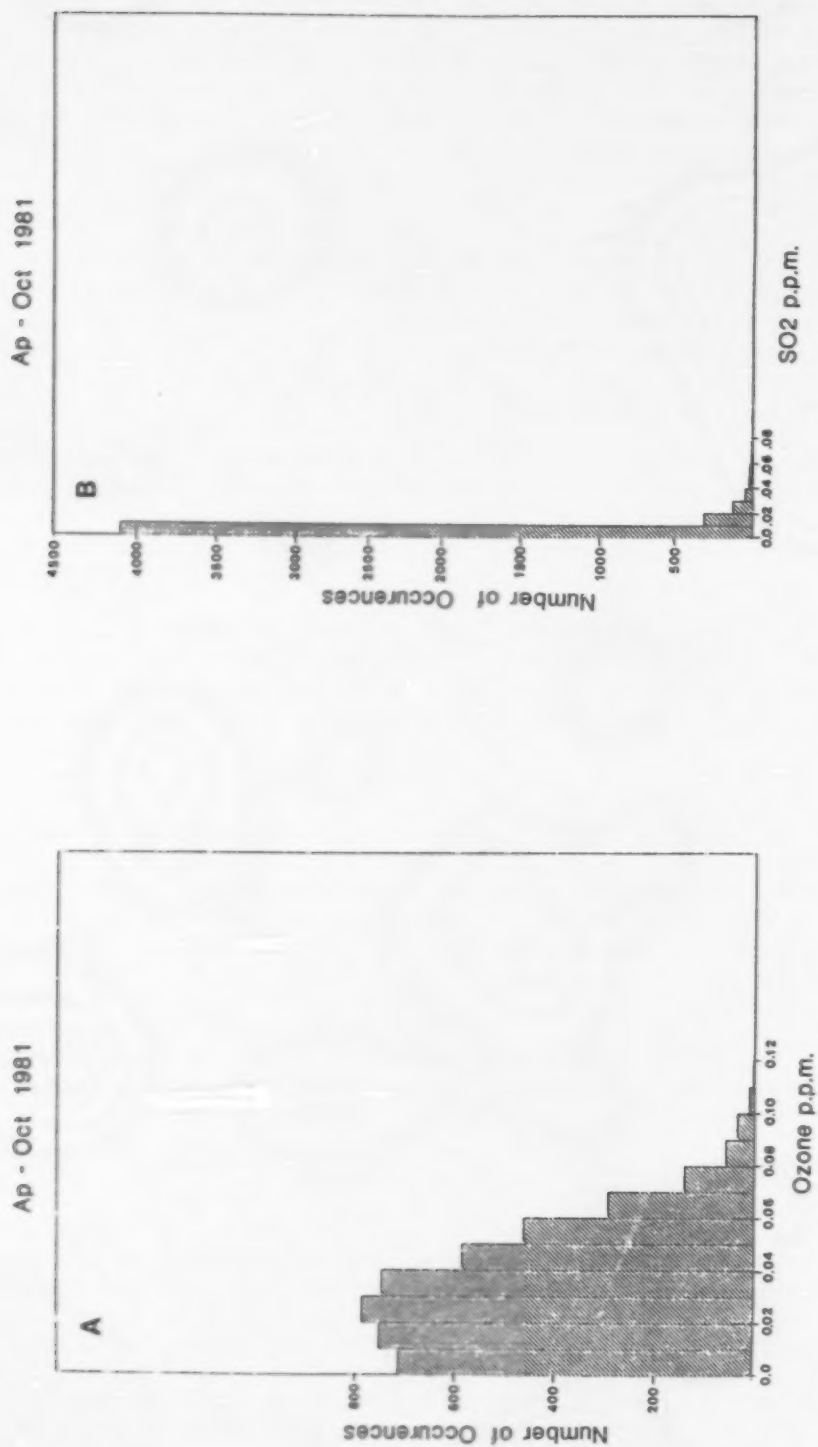


Figure V-4. Number of hourly occurrences of O₃ (A) and SO₂ (B) at specified concentration ranges from a rural site (Tarpon Springs, Florida). From Lefohn et al. (1986b).

sites under the influence of anthropogenic sources, the vast majority of the hourly SO₂ concentrations are at or near the minimum detectable level. Distributions of hourly concentrations from source and background monitoring sites in the Great Lakes states, Upper Midwest, and Southeast all indicate infrequent occurrence of hourly ozone concentrations greater than 0.05 ppm. With such infrequent occurrence of higher concentrations, the need for characterizing SO₂ concentrations regarding spatial and temporal occurrences is apparent. Unfortunately, few published characterizations of SO₂ air quality are available aside from those analyses conducted to investigate frequency of co-occurrence with other gaseous pollutants (Lefohn and Tingey, 1984; Lefohn et al., 1986b). These analyses are similar to those described previously for characterization of ozone air quality. An example of the length of an episode and the time between episodes for SO₂ air quality is given in Table V-5, indicating an average duration of hourly values of 0.10 ppm or greater of 1 hour; only three of these events occurred from April-October with an average of about 2000 hours respite time between events.

The diurnal pattern of SO₂ occurrences indicates the time during the 24-hr period at which the maxima occur most frequently; however the preponderance of concentrations near detectable limits may disguise this pattern. Two examples are shown in Figure V-5. Both display peak concentrations occurring during daylight hours.

Two patterns of seasonality are shown for the occurrence of the higher concentrations of SO₂ in Figure V-6. In one, the highest hourly occurrences over the year occurred in February, and in the other example no seasonality can be discerned for peak occurrences. Both monitoring sites were source-oriented.

Nitrogen Dioxide

The need to understand the temporal and spatial dynamics of NO₂ occurrences in regional or site-specific air quality is the same as with ozone and SO₂. At this time, however, very few of the hourly monitoring data have been characterized in other than the annual mean format. Some indication of spatial and temporal occurrence is given, at least for summer months, in the cumulative frequency distributions for a few rural sites in Table V-6. Occurrences above 40 ug/m³ are infrequent, indicating a possible episodic nature of occurrences of high concentrations, as pointed out with the two other gaseous pollutants. A seasonality of NO₂ occurrence may be important also with indications that concentrations may be 50-100% greater in the winter than in the summer (Altshuller, 1984).

Summary

Analysis and characterization of air quality and the contributing roles of the three gaseous pollutants indicate an episodic occurrence of the peaks or higher concentrations over the growing season, at least, and distinct diurnal temporal distribution of concentrations with ozone and SO₂. Any investigations of the impact of the gaseous pollutants must include these spatial and temporal characteristics of occurrence in exposure regimes designed to study impact of chronic exposure on trees. The seasonality of ozone, SO₂, and NO₂ has also been observed. This is an important element of experimental design to include

Table V-5. Average duration of SO₂ events and respite times between events indicated at Porter Co., IN (1982) (from Lefohn, personal communication).

	Event (ppm)						
	0.05	0.06	0.07	0.08	0.09	0.10	0.12
Duration (hr) of events	1.4	1.3	1.4	1.3	1.0	1.0	1.0
Number of events	26	11	8	6	5	3	1
Respite time (hr) of respites	193	414	592	829	1037	2087	---
Number of respites	25	10	7	5	4	2	---

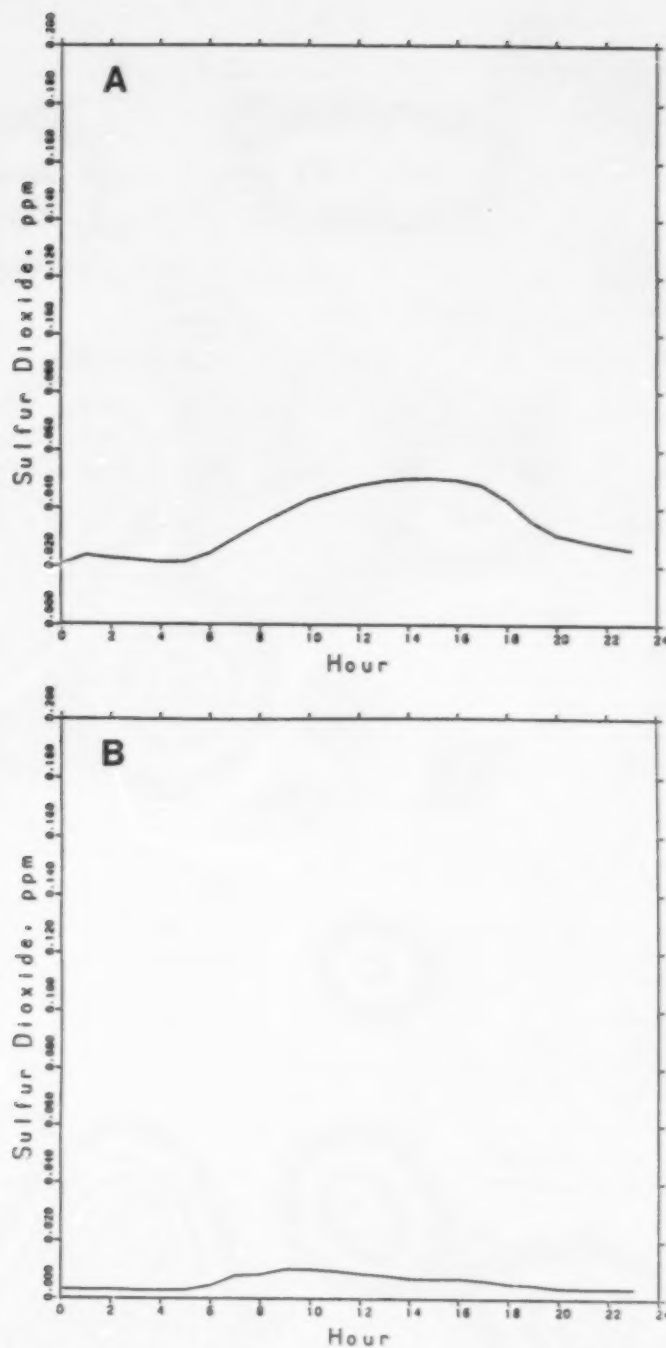


Figure V-5. Average diurnal concentration pattern for SO_2 during the growing season months. A. Columbus Co., WI (April-October 1978), and B. Porter CO., IN (May-October 1982). From Lefohn (personal communication).

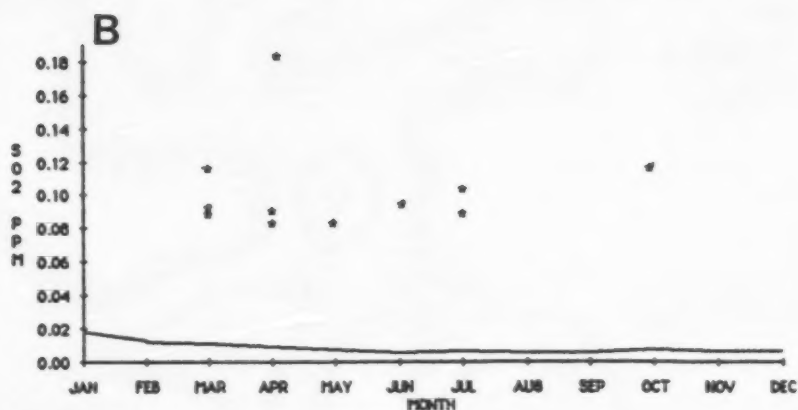
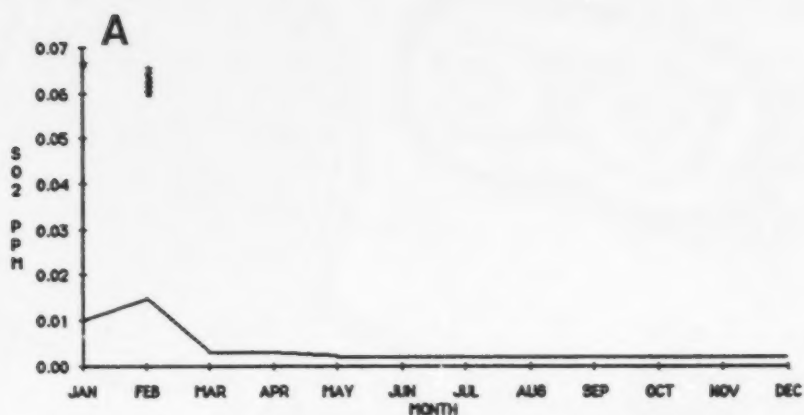


Figure V-6. Monthly average concentrations of SO₂ () with 12 highest hourly maxima over the year () to indicate the degree of seasonality in SO₂ occurrence at two source-oriented rural monitoring sites A. Columbia Co., WI; B. Porter CO., IN. From Lefohn (personal communication).

Table V-6. Cumulative frequency distribution of hourly concentrations of nitrogen dioxide at rural and suburban locations ($\mu\text{g}/\text{m}^3 \times 5.32 \times 10^{-4} = \text{ppm NO}_2$). From Altshuller (1984).

Site/Reference	Measurement Period	20 $\mu\text{g m}^{-3}$	40 $\mu\text{g m}^{-3}$	60 $\mu\text{g m}^{-3}$	80 $\mu\text{g m}^{-3}$
DuBois, PA Research Triangle Institute 1975	June-Aug. 1974	13.2	1.0	0.2	0.0
Bradford, PA Decker et al. 1976	July-Sept. 1975	2.1	0.1	0.0	0.0
McHenry, MD Research Triangle Institute 1975	June-Aug. 1974	6.9	0.2	0.1	0.0
Wooster, OH Research Triangle Institute 1975	June-Aug. 1974	23.8	6.9	1.9	0.3
McConnelsville, OH Research Triangle Institute 1975	June-Aug. 1974	5.6	0.5	0.1	0.0
Wilmington, OH Research Triangle Institute 1975	June-Aug. 1974	14.9	2.6	1.1	0.5
Creston, IA Decker et al. 1976	July-Sept. 1975	0.2	0.0	0.0	0.0
Wolf Point, MT Decker et al. 1976	July-Sept. 1975	0.4	0.0	0.0	0.0
De Ritter, LA Decker et al. 1976	July-Sept. 1975	4.8	0.3	0.0	0.0

in single pollutant studies, and even more so in pollutant combination studies. Both SO₂ and NO₂ monthly means indicate greater concentrations in the winter months, whereas ozone occurs almost exclusively March through October in the temperate regions.

Air Quality Characterization:

Wet Deposition

The trends in wet deposition of the major chemical species have been discussed in a number of reviews (e.g., Stensland, 1984) and consequently are not detailed here. Characterization of wet deposition as it relates to exposure profiles must consider the dual components of a wet deposition event: the chemical composition of the hydrometeor and the deposition rate. The large scale monitoring projects, however, have focused on the annual deposition rates and annual volume-weighted concentrations of principal ions, rather than the dynamics of the event.

Lefohn and Krupa (1984) analyzed wet deposition data from three databases for several sites over the years 1979-1981. Table V-7 and V-8 present data for 1981 as an example for annual deposition rates and volume-weighted concentrations of the principal ions. The authors were looking for deposition patterns across regions of the U.S. and changes in deposition and volume-weighted concentration of ions with season. No consistent pattern in annual volume-weighted concentration of the principal ions across the U.S. was observed; however, the seasonal deposition of hydrogen and sulfate were highest during the spring and summer and the annual deposition was less in the Western states than in the Northeast. Regional patterns of wet deposition of the important ions are shown in Figures V-7 and V-8 for the Western and Eastern states, respectively. These data provide an indication of chemical composition and at least the quantity of deposition on an annual basis for analysis of wet deposition in constructing exposure scenarios.

The frequency and duration of precipitation events and spatial and temporal trends are poorly characterized for both rain and cloud water. The most significant feature of precipitation events is the pronounced variability in space and time. For example, patterns in monthly storm events across North America are recognized as being region-specific (Hales, 1984). As a function of time (e.g., season, month), precipitation rates vary little in the Northeast from month-to-month but show pronounced variation in the Southwest and Northwest. Even more indicative of the degree of variation in incident precipitation is the observation that the coefficient of variation in deposition for a single event may be as much as 100% within a given watershed (Lindberg et al., 1977). An indication of duration and volume of events was reported by Evans (1984) where for several nonurban sites in eastern North America 88% of all rainfall events were less than 160 minutes in duration and 38-58% of all rainfall events were less than 1 mm.

The type of precipitation is also important in considering ambient air quality and impact on vegetation. For example, Rutter (1975) has described the normal hydrology of a low elevation forest with about 40 inches of rain a year

Table V-7. Annual deposition for principal ions (kg ha^{-1}) from January 1, 1981, through December 31, 1981. From Lefohn and Krupa (1984).

Site ^a	State	Ca^{2+}	Mg^{2+}	H^+	NH_4^+	Na^+	SO_4^{2-}	NO_3^-	Cl^-
Caribou	ME	1.60	0.40	0.32	1.36	0.93	19.82	7.94	1.68
Acadia NP	ME	0.94	0.83	0.40	1.58	4.83	20.44	11.02	8.58
Greenville St.	ME	1.08	0.40	0.26	1.10	1.45	15.80	7.91	2.13
Bridgton	ME	0.57	0.46	0.31	1.21	2.60	15.20	7.89	4.50
Hubbard Brook	NH	1.95	0.65	0.70	3.15	1.78	35.57	19.82	2.89
† Turners Falls	MA	0.35	0.09	0.44	1.33	0.62	14.22	10.72	1.16
* Lewes	DE	1.05	0.71	0.39	2.38	6.27	22.54	11.33	11.57
Huntington	NY	2.00	0.46	0.42	2.80	0.69	25.02	15.77	1.09
* Whiteface	NY	1.13	0.22	0.42	2.32	0.35	22.05	13.12	1.21
Knobit	NY	1.46	0.55	0.44	2.22	0.88	22.33	13.04	1.68
Stilwell lake	NY	1.42	0.86	0.59	2.14	4.54	27.82	16.47	8.51
* Brookhaven	NY	1.00	0.70	0.44	2.16	5.93	25.22	14.01	10.87
Bennett Bridge	NY	3.20	0.71	0.76	4.78	0.95	41.09	26.64	1.98
Aurora	NY	1.90	0.41	0.62	3.28	0.64	31.52	17.75	1.31
* Ithaca	NY	1.39	0.23	0.81	3.51	0.57	40.38	21.67	2.11
Jasper	NY	0.92	0.22	0.29	1.54	0.60	15.63	8.88	0.78
Chautauqua	NY	2.10	0.61	0.63	3.80	1.18	35.00	20.11	1.89
Washington Cr.	NJ	0.89	0.28	0.15	0.68	0.65	10.46	4.78	1.10
* Pennsylvania St.	PA	2.08	0.37	0.58	4.40	1.39	33.08	18.16	3.17
Leading Ridge	PA	1.81	0.52	0.66	3.08	1.16	32.99	18.44	2.30
kane	PA	1.04	0.26	0.36	1.74	0.35	17.06	12.25	1.14
Caldwell	OH	1.54	0.34	0.45	1.96	0.45	24.14	11.55	1.11
Wooster	OH	1.64	0.39	0.42	2.95	0.53	24.16	12.43	1.19
† Zanesville	OH	2.23	0.25	0.88	4.13	0.90	35.97	19.46	2.05
Delaware	OH	1.36	0.31	0.36	2.29	0.51	20.47	10.85	0.93
* Oxford	OH	1.39	0.21	0.41	2.67	0.77	22.58	11.62	1.82
* Urbana	IL	0.41	0.04	0.16	1.37	0.14	10.56	5.22	0.39
Calibration									
Spooner	WI	1.73	0.37	0.07	3.53	0.59	12.11	8.59	0.68
Mead	NE	2.58	0.40	0.05	3.71	0.72	11.59	8.11	0.97
Huron	SD	1.37	0.30	0.01	2.27	0.26	4.62	4.64	0.50
Manitou	CO	1.38	0.27	0.07	0.91	0.34	6.24	5.80	0.46
Hopland	CA	0.52	0.59	0.04	0.73	3.99	3.41	2.61	7.42
Schmidt Farm	OR	0.96	0.53	0.02	0.94	2.89	4.18	1.75	5.08
Olympic NP	WA	2.22	2.72	0.12	0.61	22.56	12.87	2.74	42.03

^a Database from NADP except those marked (*) are from MAP3S and (†) are from EPRI.

Table V-8. Annual volume weighted concentration values for principal ions (ueq l⁻¹) from January 1, 1981, through December 31, 1981. From Lefohn and Krupa (1984).

Site ^a	State	Ca ²⁺	Mg ²⁺	H ⁺	NH ₄ ⁺	Na ⁺	SO ₄ ²⁻	NO ₃ ⁻	Cl ⁻
Caribou	ME	7.30	3.08	31.06	7.21	3.62	39.81	11.97	4.38
Acadia NP	ME	5.17	7.51	43.89	9.53	22.78	46.68	19.13	26.14
Greenville St.	ME	4.93	2.91	23.54	5.51	5.45	29.73	11.50	5.04
Bridgton	ME	4.01	4.34	41.13	9.11	11.79	41.83	17.71	12.96
Hubbard Brook	NH	6.29	3.47	45.66	11.56	5.05	48.44	20.44	5.31
† Turners Falls	MA	6.16	2.67	82.25	15.77	7.60	54.54	39.61	8.90
* Lewes	DE	6.19	6.91	45.99	15.54	32.02	55.12	21.45	38.25
Huntington	NY	8.16	3.33	41.22	13.08	2.81	48.80	22.83	2.82
* Whiteface	NY	5.03	1.64	37.63	11.40	1.34	40.71	18.75	3.01
Knobit	NY	8.58	5.36	51.70	14.26	4.57	54.89	25.57	5.39
Stilwell lake	NY	8.21	7.15	68.65	14.21	17.74	67.51	30.87	21.43
* Brookhaven	NY	5.17	5.82	45.54	12.53	25.71	54.26	23.48	30.36
Bennett Bridge	NY	11.50	4.41	61.04	20.24	3.09	67.13	33.70	4.24
Aurora	NY	11.66	4.12	80.27	22.80	3.23	84.57	35.79	4.41
* Ithaca	NY	6.14	1.69	71.50	17.19	2.21	74.26	30.85	5.24
Jasper	NY	10.06	3.83	50.63	15.22	4.43	60.24	25.93	3.80
Chautauqua	NY	10.81	4.47	69.15	21.73	4.73	78.49	33.14	5.07
Washington Cr.	NJ	9.66	5.21	38.72	9.78	7.61	53.22	18.77	8.51
* Pennsylvania St.	PA	10.46	3.10	58.35	24.65	6.11	69.50	29.53	9.00
Leading Ridge	PA	9.02	4.29	68.55	17.79	5.15	71.13	30.28	6.62
kane	PA	8.83	3.41	58.88	16.07	3.25	60.16	31.86	5.05
Caldwell	OH	14.07	4.71	80.57	19.22	5.51	91.74	31.66	6.01
Wooster	OH	12.37	4.87	59.45	23.71	3.68	72.60	29.27	5.20
† Zanesville	OH	14.14	3.24	79.94	24.37	6.93	71.18	34.00	7.42
Delaware	OH	10.76	3.87	60.19	20.34	3.64	69.60	28.68	4.15
* Oxford	OH	9.21	2.31	54.87	19.55	4.41	62.10	24.73	6.75
* Urbana	IL	5.94	0.91	47.81	22.20	1.77	64.13	24.55	3.20
Spooner	WI	13.40	4.80	10.64	30.48	3.77	40.09	21.07	2.64
Mead	NE	19.05	4.98	6.66	30.94	4.53	26.49	19.55	3.85
Huron	SD	18.71	6.91	2.07	40.07	3.28	29.83	23.21	4.27
Manitou	CO	17.01	5.82	17.33	13.20	3.69	33.83	24.36	3.37
Hopland	CA	2.46	4.73	3.82	3.90	16.84	6.73	3.92	20.35
Schmidt Farm	OR	5.84	5.56	2.72	6.10	16.55	10.68	3.36	19.18
Olympic NP	WA	4.02	7.70	4.19	1.18	33.79	9.60	1.66	41.18

^a Database from NADP except those marked (*) are from MAP3S and (†) are from EPRI.

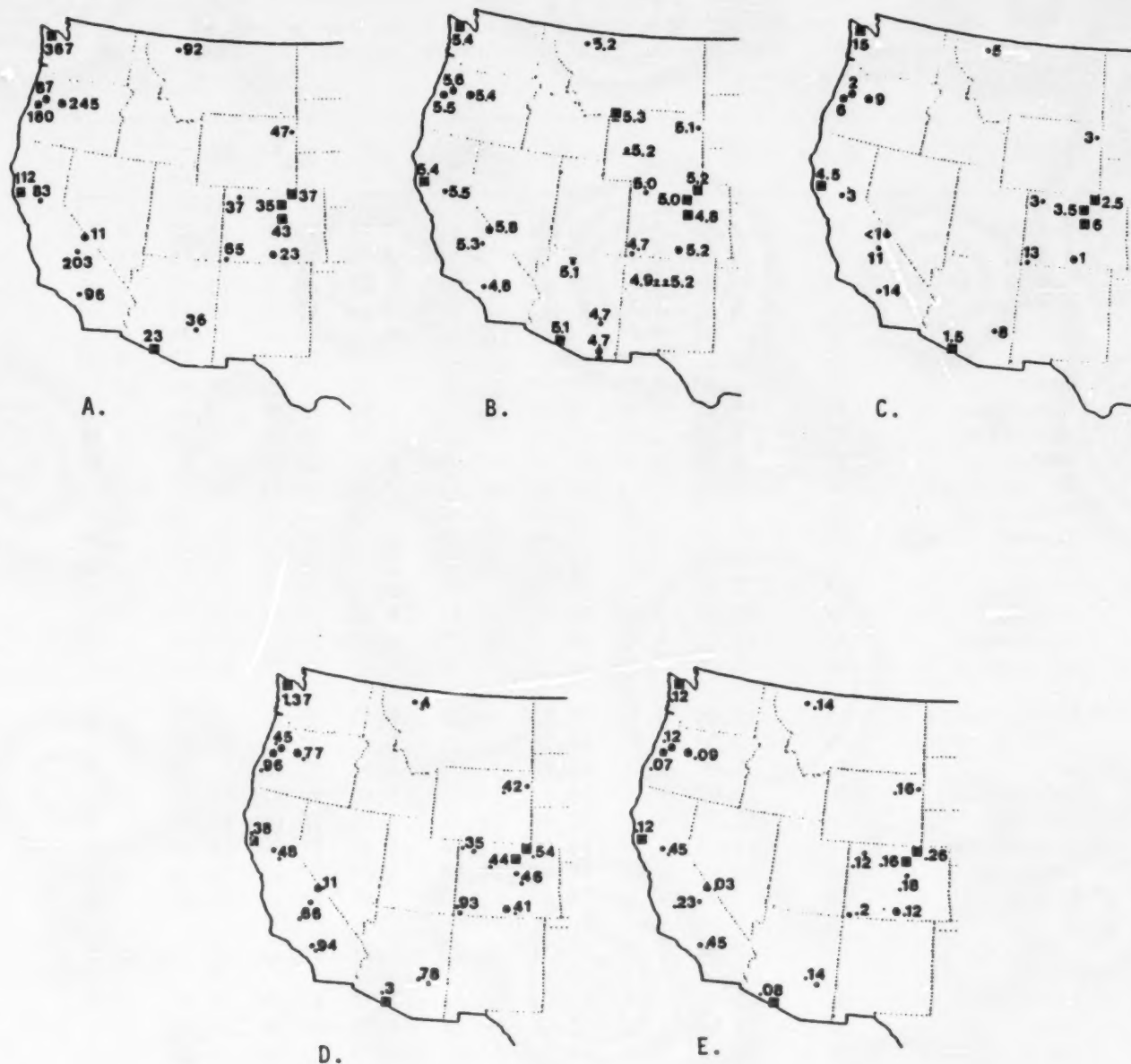


Figure V-7. Mean annual wet deposition at western NADP sites. A. Precipitation (cm), B. pH (volume weighted mean), C. H^+ ($g\ m^{-2}$), D. SO_4^{2-} ($g\ m^{-2}$), E. $NO_3^- - N + NH_4^+ - N$ ($g\ m^{-2}$). = 1981, = 1982, = 1981-82. = data not quality assured by NADP. From Olsen and Watson (1984).

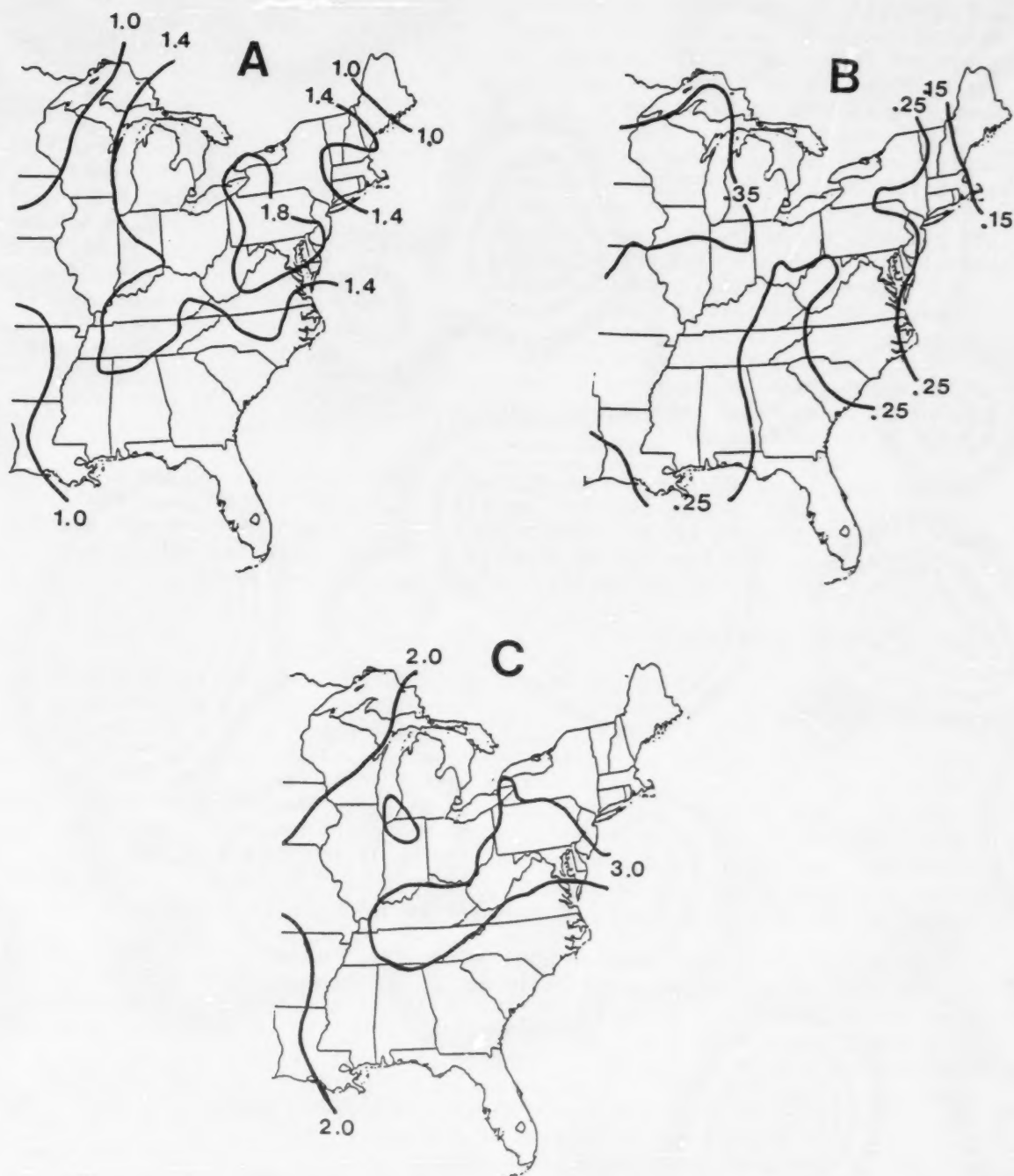


Figure V-8. Wet deposition in the eastern U.S. for 1982. All values g m^{-2} . A. NO_3^- , B. NH_4^+ , C. SO_4^{2-} . From NADP (1985).

and approximately 20 inches evaporation. For this type of area, mist and fog make up less than 2% of the water input. In contrast, Lovett et al. (1982) suggested from models that fog water deposition accounts for almost half of the water input to high elevation fir forest in New Hampshire. In the Green Mountains of Vermont, Vogelmann et al. (1968) estimated that 70% of the precipitation is via fog water. Analysis of existing data would suggest that the deposition in fog water for mountaintops could equal the deposition in rainfall and dry deposition combined (R. Bradow, personal communication); thus analysis of ambient air quality for mountainous forests should include considerations of the dynamics and chemical composition of fog. Monitoring of fog deposition has been limited, but programs are beginning (R. Bradow, personal communication, S. E. Lindberg, personal communication). In addition to chemical analysis of fog water for principal ion concentration, the temporal and spatial nature of the events, particle size distribution, wind speed, etc. are necessary to collect in monitoring programs (Bradow, personal communication). An example of fog water chemistry is shown in Table V-9, and indicates the much greater deposition of sulfate, nitrate, and ammonium ions by fog than by bulk precipitation in the White Mountain area.

Air quality in a given region may have elements of both wet and dry deposition as well as more than one important component within the overall general description of wet or dry deposition. Characterization of air quality for pollutant combinations has received only limited attention and yet is undoubtedly important in evaluating air quality impact on forests.

Co-Occurrence of Pollutants

Most studies concerning the impact of pollutant combinations on plants have assumed that there are frequent periods of pollutant co-occurrence. However, this assumption is not supported by analyses of actual air quality data. Only a few studies have attempted to describe the co-occurrence of pollutants in the atmosphere and related this information to a biological viewpoint. This lack of characterization data is a consequence of both limited ambient monitoring for multiple pollutants at a given site and also of the conception that the air must contain multiple pollutants just by its very nature and therefore it is not necessary to resort to actual air quality data to describe the joint occurrence of multiple pollutants.

Although air pollutants have been co-monitored at only a few sites, these provide sufficient indication of pollutant co-occurrence. For example, in a study using data from SAROAD (1981), TVA (1978 to 1982) and EPRI SURE/ERAQS (1978 and 1979), Lefohn and Tingey (1984) found 91, 124, and 147 site-years of data available in which the pollutant pairs, SO_2/NO_2 , O_3/SO_2 , and O_3/NO_2 , respectively, were monitored simultaneously. The authors defined pollutant co-occurrence as the simultaneous occurrence of two pollutants with hourly mean concentrations of 0.05 ppm or greater of each gas. Based on this criterion, episodes of pollutant co-occurrence usually lasted only a few hours per week and the interval between episodes was generally large (weeks to sometimes months). Most monitoring sites experienced fewer than 9 hours of pollutant co-occurrence, for any of the three pairs of pollutants during the 5-month growing season (May to September).

Table V-9. Annual deposition in cloud capture and bulk precipitation. Adapted from Lovett et al. (1982) (Bradow, personal communication).

Ion	Cloud Deposition kg/ha-yr	Bulk Precipitation kg/ha-yr	Percentage from Clouds
H^+	2.4	1.5	62
NH_4^+ (as N)	13.4	3.4	80
Na^+	5.8	1.7	77
K^+	3.3	2.1	61
SO_4^{--} (as S)	91.9	21.5	81
NO_3^{--} (as N)	22.9	5.3	81

Analysis of data from a portion of the Ohio River Valley that contained four coal-fired power plants supports the conclusion that the co-occurrence of SO_2 and NO_x was small (Jacobson and McManus, 1985). Using minimum concentrations of 0.05 ppm and $58 \mu\text{g m}^{-3}$ for SO_2 and NO_x , respectively, the authors showed that the two gases occurred jointly for less than 1% of the total hours monitored. Analysis of air quality data from central London, during the heating season (January through March), also found that the co-occurrence of SO_2 and NO_2 was small, less than 1% of the time (Lane and Bell, 1984a). The occurrences of SO_2 and NO were positively correlated; simultaneous occurrences were measured between 2 and 4% of the monitoring period when a minimum concentration of 0.05 ppm for each gas was used.

The analyses of Lefohn and Tingey were criticized because they: (1) used only one year (1981) of SAROAD data; (2) included a large number of non-rural sites; (3) used a relatively high (0.05 ppm) concentration threshold to determine pollutant co-occurrence; and (4) failed to consider the importance of possible sequential pollutant occurrence within the same day. In response to these issues, Lefohn et al. (1986b) have extended the analysis of pollutant co-occurrences for rural sites to consider both simultaneous and sequential occurrences using a lower (0.03 ppm) concentration threshold and several years of data. Even using the lower concentration to define a simultaneous pollutant co-occurrence did not change the conclusion that simultaneous occurrences were rare. For any of the two pollutant pairs, the majority of sites experienced fewer than 10 to 20 hours of simultaneous occurrence during a 5-month period (May to September; 3672 hours). In addition, sequential occurrences, when both pollutants exceeded the minimum concentration (0.03 ppm) within the same day, were also rare events during the 5-month (153 days) period. The majority of the sites experienced fewer than 10 to 20 days of pollutant co-occurrence. The low numbers of simultaneous and sequential occurrences were observed in all five years (1978 to 1982) of data analyzed. Although not well characterized from a biological perspective, seasonal patterns of co-occurrence should be considered, especially in relation to possible effects on long-lived perennial species. For example, ozone tends to be elevated during the second and third quarters of the year while in many areas sulfur dioxide would display its highest concentrations during the first and/or fourth quarters of the year (e.g., Figure V-6a), but at other sites (Figure V-6b) there is no apparent seasonality in the occurrence of high concentrations of sulfur dioxide.

While the temporal and chemical dynamics of individual dry or wet deposition events are recognized as important parameters to characterize in evaluating the influence of changes in air chemistry on the productivity of vegetation, the temporal and chemical relationships of the combination of sequential dry and wet deposition events is unclear. It is hypothesized that the duration and chemistry of the dry deposition period preceding a wet deposition event are important because: (1) the potential for interactive effects of sequential pollutant exposures (e.g., dry deposition of ozone followed by the interception of acidic cloud water; Krause et al., 1984; Shriner et al., 1986); and (2) the potential hydration on the leaf surface of previously dry-deposited pollutants (e.g., sulfur dioxide) which would enhance their mobility and phytotoxicity (Hoffman et al., 1980). In a reciprocal fashion, a wet deposition event immediately prior to a period of elevated levels of a gaseous pollutant might predispose the plant to injury through a modification of the plant's water

relations. It is likely that the more ephemeral the precipitation event (e.g., brief rain events, cloud water interception), the more concentrated (in the liquid phase on the leaf surface) will be the solubilized dry-deposited pollutants (Evans, 1984). Consequently, on a theoretical basis, the temporal and chemical features of sequential periods of wet and dry deposition are likely to be important features of air quality characterization. However, the temporal association of dry and wet deposition events is not well characterized. This lack is, in part, the result of different temporal measurement scales for measuring dry and wet deposition and also the lack of co-located monitors for both types of deposition. Also, scientists have only recently become aware of the importance of conducting studies that include both wet and dry deposition. Using the SAROAD air quality data base and wet deposition data from NADP and EPRI, Lefohn and Benedict (1983) concluded there was a potential for periods of elevated ozone and acidic precipitation to occur over the same geographic area during the growing season. Similarly, from an analysis of ozone concentrations and wet deposition events, Taylor and Norby (1985) concluded that the region of the eastern United States experiencing the highest annual rainfall acidity (pH 4.2) also experienced elevated ozone exposures.

An example of the potential for sequential exposures to elevated levels of ozone and wet deposition events is provided by a review of data for a deciduous forest in Tennessee. In this area there are generally five cycles of "defined periods of dry and wet deposition" per month during the growing season, with a dry period defined as being > 2 days in duration (G. M. Lovett, personal communication). During the same time interval, periods of elevated ozone levels commonly average two-to-three per month with each event averaging two-to-three days (Taylor and Norby, 1985). Thus, at a maximum, only three of the wet deposition periods per month are likely to be preceded by a dry deposition period of elevated ozone. This sequential occurrence is likely to be even less frequent both earlier and later in the growing season as the frequency of sustained elevated levels of ozone declines. Because this analysis is based upon site- and year-specific data, the pattern should not be generalized indiscriminately, but it does indicate that the frequency of sequential exposures involving wet and dry deposition is not likely to be much greater than three or four events per month.

C. Exposure Regimes for Determining the Effects of Deposition on Plants

The methods for determining the effects of wet and dry deposition on vegetation are as diverse as the disciplines participating in the investigations. The assessment of effects involves dry and wet deposition monitoring, exposure methodologies, and statistics as well as various biological and physical sciences which provide the tools for the diagnosis and evaluation of air pollution impacts. It is obvious that different experimental objectives require different experimental and exposure methodologies.

To establish and determine the effects of air pollutants on plants several basic criteria must be satisfied: (1) the conditions (climatic, edaphic, exposure, etc.) must approximate or be representative of those in the area of interest; (2) a relationship must be established between the cause and the resultant effect; (3) the exposure-response relationships must be quantified. To satisfy these criteria, a range of experimental approaches with several levels of environmental control is required.

Exposure Regimes

In studying the effects of dry and wet deposition on plants a number of exposure regimes has been used, ranging from artificial "square wave" exposures to highly sophisticated simulations of ambient conditions. The types of exposure regimes selected for use in plant response studies have been influenced by the specific objectives of the individual investigators. For example, "square wave" exposure regimes frequently have been used to conduct initial or exploratory studies of plant response to a new pollutant or for screening a large number of plants for their relative sensitivity. Sophisticated simulations of ambient air quality distributions have been used to investigate the components of an exposure that are most important for influencing plant response and to provide a realistic representation of the exposure so that the results are readily extrapolated to ambient conditions.

In addition to changing experimental objectives, exposure regimes have become more sophisticated as technology and equipment have improved, resulting in decreased cost over time. These advanced control systems are easier to operate and the expertise to manage them is readily available. As scientists better understand the relationship between plant response and the temporal and spatial patterns of pollutant exposure, there is the companion need to understand the characteristics of an exposure that impairs plant performance. This awareness has fostered development of exposure regimes representative of ambient air quality and that can also be modified to test specific biological hypotheses.

The types of exposure regimes that have been used in dry and wet deposition studies will be reviewed separately. The review is not comprehensive, but briefly describes general types of exposure regimes, listing some of the strengths and limitations of each. References are not provided for regimes with historic use, but are given for some of the newer protocols or ones that are not widely cited in the literature.

1. Dry (Gaseous) Deposition

The types of exposure regimes used in dry deposition studies can generally be separated into two classes, artificial and episodic. The distinguishing factor between the two is whether there was an overt attempt to represent the temporal patterns of ambient pollutant concentrations.

a. Artificial Exposure Regimes

Although there are many types of artificial regimes they all have the same characteristic: the regime was not developed to represent the temporal variation of ambient exposures but to meet other specific objectives; and the concentrations and exposure durations are frequently much higher and longer than ambient. Many studies have used "square wave" exposure regimes, of either short or long durations, i.e., the concentration was approximately constant for the duration of the study. Most investigation into the effects of pollutant combinations have also used artificial exposure regimes because the frequency of pollutant co-occurrence and the duration of exposure were much larger than those found in the ambient air.

Long-term artificial exposure regimes are not well suited for studies designed to determine the effects of air pollutant(s) on plant growth or yield because the exposures are not representative of ambient. Consequently, it is difficult to relate the results to ambient conditions. However, for some physiological and biochemical studies, preliminary work on a problem or for cases where there is insufficient data to develop a more realistic regime, they may prove useful.

b. Episodic Exposure Regimes

This class of regimes range from actual ambient to those that simulate ambient. The distinguishing characteristic is an overt attempt by the scientist to represent the actual temporal concentration and frequency variations that exist in ambient air quality data. Some types of episodic exposures have been used for years while others are recently developed. The episodic regimes can be segregated into three classes: ambient, modified ambient, and simulated ambient.

(1) Ambient Profiles

These regimes include comparisons of filtered and unfiltered air in various types of chambers or air exclusion systems as well as placement of plots at varying locations along a pollution gradient, usually near a point source of pollution.

The comparison of the effects of filtered and unfiltered air on plants permits the determination of the potential impact of the current pollution load on plant performance as a consequence of the actual air quality at the location of the study. However, because only a control and one treatment level are used

it is not possible to construct exposure-response functions; inferences cannot be made concerning what would happen if the air quality either improved or deteriorated. Also the data are site-specific preventing inference to the region or beyond.

Placement of plots along a pollution gradient permits the assessment of plant performance under the actual air quality and environmental conditions that exist. But unlike the filtered-unfiltered comparisons, a range of pollutant(s) concentrations is used and thus exposure-response functions can be generated. An ambient gradient has been used to determine the effects of sulfur dioxide (e.g., Guderian and Stratmann, 1962a,b) and ozone (e.g., Oshima et al., 1976) on plant growth and yield. However, unless the pollution gradient is steep or exists over an area of uniform climatic and edaphic conditions, the influence of these factors can confound the results. Other factors may also differ along the gradient which may cause additional problems.

(2) Modified Ambient Regimes

These approaches are, in reality, extensions of the ambient exposure methodologies using filtered- and unfiltered-air but with an added design element, attempting to create a range of exposure conditions so that exposure-response functions can be developed. This approach is directly coupled to the current ambient concentrations and consequently the same exposures cannot be repeated at different locations or under different environmental conditions. The modified ambient exposures have been used extensively to assess the impact of air pollutants on plant growth and yield.

There are two basic approaches to creating a range of conditions so that exposure-response functions can be developed. In the first, additional ozone is added, as either a range of fixed concentrations or proportions of ambient concentration, to create a range of exposure levels (e.g., Heck et al., 1982; Temple et al., 1985) as was done in the National Crop Loss Assessment Network (NCLAN) Program. The ozone-addition approach can create exposures at the ambient level and higher, but not for exposures below the current ambient. No studies have been published to determine if the pollutant frequency distributions resulting from the ozone-addition approach preserve a frequency distribution similar to that found in the ambient air. A second approach has used differential air filtration in which varying proportions of charcoal-filtered air and unfiltered air are mixed to produce a range of pollutant concentrations (e.g., Oshima, 1978; McCool et al., 1986). This approach can only produce concentrations up to the current ambient level, and is most appropriate for areas that have a relatively high natural pollution load. To develop a range of exposure conditions below and above the current levels using the modified ambient approach, it would be necessary to combine both the pollutant-addition and the differential-filtration methods.

(3) Simulated Ambient Regimes

Although various methods have been used, all of the simulated ambient approaches take actual ambient air quality data to create exposure regimes that simulate the observed temporal concentration changes.

The simplest approach is to obtain air quality data for a given site and time period and then reproduce that exposure under defined experimental conditions (e.g., McLaughlin *et al.*, 1976). This method works well to determine if a specific exposure condition is phytotoxic, but the resultant data are source- or site-specific, limiting extrapolation.

The statistical approaches have analyzed and mathematically characterized ambient air quality. The resultant mathematical parameters were used in a statistical model to construct exposure regimes. The log-normal statistical parameters that characterized ambient sulfur dioxide concentrations were used in a computer program to stochastically create a range of exposure concentrations to study the effects of sulfur dioxide on crops (Male, 1982; Male *et al.*, 1983).

Pragmatic approaches have also been used which mathematically characterize the ambient air to develop regimes that simulate temporal variations of ambient exposures. Some use actual air monitoring data from one to several sites for characterization (Lane and Bell, 1984a, 1984b; Hogsett *et al.*, 1985; Lefohn *et al.*, 1986a, 1986b), while others have used predicted hourly concentrations generated by pollutant dispersion models (Laurence and Kohut, 1984). Rather than attempting to determine the specific statistical parameters of the distribution, the pragmatic approaches use statistical correlations, frequencies of occurrence, duration of and between events and other "rules of thumb" to generate regimes that maintain the same statistical properties as the original air quality data used to develop them. The air quality that is characterized in the pragmatic approach can represent a site or a range of sites making up a region of interest for one or more years. The broader data set allows development of average characteristics and variations thus providing opportunity for wider extrapolation of the data.

The simulated ambient approaches can be used to develop exposure-response functions for plants exposed to various air pollutants. The effects of pollutant combinations on plants can be determined using realistic frequencies of pollutant co-occurrence. Because the regimes are mathematical representations of ambient conditions they can be used to investigate the components of a pollutant exposure that influence plant response. Simulated ambient exposures provide the experimental flexibility necessary to adapt to changing biological objectives. Despite their obvious realism and experimental advantages, the simulated ambient exposures require sufficient ambient air quality data to characterize the ambient concentrations and their temporal variations before exposure regimes can be developed.

The simulated exposures are realistic representations of the ambient air because they maintain the temporal concentration variation and the statistical properties of actual exposures. These characteristics permit the resultant data to be extrapolated to ambient conditions, especially if the environmental

conditions are realistic. Both the statistical and pragmatic approaches permit the generation of exposure regimes that are characteristic of a given area, not just a specific site. These approaches are well-suited for creating a range of exposure concentrations necessary for the development of exposure-response functions and for long-term studies on plant growth. The simulated ambient regimes can be reproduced over time, at several locations and under a range of environmental and edaphic conditions to yield the required replication of a response over time and space. This replication is necessary to establish cause-effect relationships required to determine if airborne pollutants are impacting forests.

2. Wet Deposition

Exposure regimes for wet deposition are potentially more complicated than those for dry deposition studies. They must recognize the dual nature of a wet deposition event which includes both the chemical composition of the hydro-meteor (which can take several forms) and its deposition rate. Experimental protocols used to investigate the impact of wet deposition on plants have attempted to (1) determine relative sensitivity using various biological indicators; (2) determine the mode of action; and (3) develop exposure-response functions which mathematically relate plant response to wet deposition exposure. The term "exposure-response" suggests a univariate association between exposure and plant response. However, a wet deposition event includes several different parameters; consequently complex factorial or multivariate analyses may be required to define the "exposure-response" relationships adequately.

Studies of wet deposition impacts on vegetation have been conducted in both controlled environments (e.g., greenhouse chambers, controlled environment chambers) and under field conditions. Controlled environment studies are useful indicators of potential impacts and permit the study of effects not readily measured under less controlled field conditions. Plant variability in controlled environments is frequently less, allowing the detection of smaller differences; these type studies are well suited for physiological and biochemical studies as well as short-term (a few days) screening studies. However, because the cultural and environmental conditions are substantially different from those in the ambient, it is difficult to extrapolate the results to the field, especially for long term growth and yield studies. Plants grown in controlled environments are more sensitive to acid rain injury than field grown plants (Irving, 1984). In most controlled environment studies the total deposition of H^+ , SO_4^{2-} , and NO_3^- is greater than in field studies because of a larger number of exposures and/or a higher deposition rate.

In field studies, the environmental conditions are the same as or similar to ambient conditions. Consequently the data are site-, or at most, region-specific, and it is difficult to extrapolate the data to other climatic or regional conditions. Because the conditions are less controlled, there is usually greater plant variability than in controlled environment studies. To manage for this variation and obtain reliable data requires large numbers of plants as well as studies conducted over several years. Although labor and cost intensive, field studies are often the only way to obtain the necessary type of data. Another frequent problem with field studies is the lack of a

comparable unpolluted control. In many regions it is necessary not only to control or exclude the wet deposition events but also to control other pollutants (gaseous or dry) that are not a part of the study so they do not confound the experimental results.

In the field, wet deposition exposure regimes can be divided into two classes: those that receive wet deposition in addition to ambient rain events and those in which the ambient wet deposition is excluded and a simulated wet deposition is applied.

The addition of wet deposition to ambient rain requires relatively simple equipment and usually induces little or no change in the environment around the plants. The plants are exposed to the ambient array of wet and dry deposition types. The approach cannot determine vegetation effects at or below the current ambient deposition level; also the crops receive an unusually large amount of water and larger depositions of H^+ , SO_4^{2-} , and NO_3^- than would be typical for the area.

The exclusion of ambient wet deposition and the subsequent simulation of wet deposition permits the development of exposure-response functions and the determination of potential impacts both above and below the current ambient wet deposition level. However, for the data to be most useful, the study must simulate not only the wet deposition chemistry and hydrometeor characteristics, but also ensure that the environmental conditions during a wet deposition event are similar to ambient conditions. The methods for excluding and simulating wet deposition are relatively expensive and may alter the environmental conditions from the ambient. Because wet deposition is excluded, the method must provide additional water to the vegetation to compensate for the loss of ambient rainfall.

Exposure protocols used to investigate the influence of wet deposition on vegetation are defined by the combination of precipitation chemistry and the multiple aspects of the hydrometeor's deposition rate which include deposition rate (cm/h), frequency and duration of events. The temporal dynamics of these dual features are important to consider in developing exposure regimes. One conspicuous feature of wet deposition studies is the extreme diversity among the experimental designs in the selection of chemical and depositional properties. The chief explanations for this diversity are the pronounced spatial (e.g., continent, region, elevation) and temporal (e.g., season, year) variation in precipitation chemistry and climatology and the multiple forms in which precipitation can occur.

To simulate the chemistry of the hydrometeor, most studies have used published records of ambient rainfall or cloud water chemistry to develop their exposure regimes. However, the extent of the mimicry is commonly restricted to selected aspects of the ambient precipitation. For example, the most common simulant specification is acidity, achieved through the addition of sulfuric and/or nitric acids. While many designs maintain a realistic sulfuric-to-nitric acid ratio in achieving the desired hydrogen ion concentration, this ratio specification is not always appreciated. In many of the regimes, the chemical characterization has considered only the sulfuric/nitric ratio or the hydrogen ion concentration without taking into account other cations and anions

that are common components of precipitation (e.g., geologic or anthropogenic cations and anions, including trace elements, heavy metals, organics, and free radical species). This aspect is essential when considering cloud water/mist because concentrations of multiple cations and anions tend to be enriched in cloud water relative to rainfall in the same region.

Most of the exposure protocols have maintained a constant ("square wave") chemical profile (based on growing season average concentrations for the individual ions for the area), for the duration of the wet deposition event with the same event repeated throughout the duration of the study. However, in most natural rain events the hydrometeors show an initial enrichment in sulfates, nitrates, and hydrogen ions compared to droplets occurring later in the event. Although no studies have investigated the importance of "peak" events versus total deposition, studies have shown that plant response is different when the rain chemistry varies from event to event rather than being the same all the time (Irving, 1984). Only a few studies have attempted to increase the pH during a rain event (e.g., Kelly *et al.*, 1984) as occurs in nature. Johnston *et al.* (1982) reported that the growth of bush beans was reduced more when exposed to acidic rain (pH 3.2) in which the hydrogen ion concentration varied during the event than in plants exposed to a constant concentration of the same average pH.

The most common method of varying the exposure regime is through changes in the depositional features of the hydrometeor. The physical parameters used to characterize the hydrometeor encompass the rate/intensity of the event (cm/h), event duration (h), number of events/week, duration of the time between precipitation events, and the periodicity of the events. Most protocols establish some combination of rate, duration, and number of events per week to achieve a predetermined hydrometeor or chemical deposition rate which is commonly selected based on ambient climatological or chemical data. This predetermined specification is typically a multiple year or seasonal average and thus may not convey many of the stochastic or rhythmic features of ambient wet deposition. As with the chemical aspects of wet deposition protocols, most of the physical features can be characterized as being "square wave". Thus, the duration, intensity, number of events per week, and the periodicity in most exposure protocols are invariant, with the only variable being the duration of the inter-event period. This feature is descriptive of most controlled and field exposure protocols.

Exposure protocols with respect to cloud water/mist interception have additional features that separate them from typical rain deposition studies. The concentrations of various ions in cloud water/mist are typically enriched with the same concentrations of multiple cations and anions as the region's rainfall. Also, cloud water deposition occurs predominately to the foliage rather than the soil. Most experimental protocols for cloud water/mist studies qualitatively (rather than quantitatively) characterize deposition according to the degree of surface wetness or immersions by the event. Commonly, the degree of wetness is invariant in repeated exposures so that the chemical deposition rate is constant within and among events.

Studies should also consider the area being simulated. For example, it is estimated that high elevation forests are immersed in advective clouds as much as 50% of the time (annual average). With a decrease in elevation, the percent of time that the foliage is wetted by clouds declines to near zero.

VI. Recommendations

A. Workshop Guidelines

The workshop was structured to obtain a collective judgment on recommendations for standardizing pollutant exposure systems (both wet and dry) and exposure protocols for the Forest Response Program of the National Acid Precipitation Assessment Program. The participants were working either directly with pollutant exposures as biologists or engineers, or indirectly in the forest sciences, contributing the perspective of tree growth and assessment. The workshop was informal and interactive; the objective was to achieve a consensus on the recommended level of standardization of exposure studies with forest species. Small, interactive groups explored and discussed recommendations for standardization within the three categories: (1) dry deposition exposure systems; (2) wet deposition exposure systems; and (3) deposition exposure regimes. Participants were asked to answer:

1. What is the minimum level of standardization of exposure studies (wet and dry deposition) to pursue?
2. Should that level of standardization be applied to specific equipment/facility or should the level of standardization be applied to performance characteristic(s)?
3. What is the recommended standard of exposure system and protocol for specific research objectives?

The preceding chapters reviewed existing exposure systems and evaluated them with criteria designed to determine the system's ability to provide reproducible experimental conditions; discussed the strengths and limitations of the various types of exposure systems; and identified approaches used to develop exposure regimes for fumigation studies. Decisions recommending a standardized system and/or standard performance characteristic, emphasize the obvious need to consider the research or biological objective and the practices required to work with tree species. This caveat pertains to the previous draft document provided the participants and to the recommendations offered here.

The objective of the workshop was a consensus recommendations for standardizing exposure systems and protocols in forestry species studies. As a clear guideline to achieve this end and recognizing the need to consider the biological objectives, the participants focused their discussion on the following generic biological objectives, tempered with concern for the specific requirements of forest species:

1. Multi-year (2-5 years) growth/physiological studies
 - A. Seasonal exposures
 - B. Year-round exposures
 - C. Climatic and biotic interaction with exposure (e.g., drought stress, nutrient stress, pathogens)

2. Short-term (< 2 years) growth/physiological studies

3. Gas-exchange/pollutant uptake studies

In the above categories, the age or size of plant material, the species (deciduous or coniferous), and pollutant type will also influence the recommendation(s).

For each of these broad objectives and for the type of pollutant, the participants were instructed to answer the following questions for each of the generic objectives:

1. Chambered versus non-chambered systems?
2. Type of chambered system? or performance characteristic?
3. Type of non-chambered system? or performance characteristic(s)?
4. Standardization of the monitoring needs from equipment perspective as well as sampling frequency.
5. For each generic research objective, address the appropriateness of standardized exposure regimes. If appropriate, what would the standardized exposure regime consist of and how should it be developed.

It was important to acknowledge that there might not be unanimity for certain standardization recommendations, and that was acceptable. Dissenting views would be incorporated into the final recommendations if there were valid scientific reasons.

B. General Recommendations for Exposure System

There was workshop consensus that certain elements of exposure studies should be standardized and these are presented in Table VI-1. The first five items in Table VI-1 were identified as those to be standardized within and between programs; the details should be specifically determined in the individual quality assurance programs. Certain points, however, were emphasized: (1) the need to use EPA-approved monitoring equipment; (2) ensure that sampling frequency is sufficient to characterize hourly mean concentration values, perhaps utilizing time-series models to determine the appropriate sample size; and (3) ensure that gaps in monitoring data be handled the same at all research sites. In developing quality assurance programs, the primary objective of useful air quality data from all research sites must be paramount. Acquisition of micrometeorology should be of high priority in both chambered and non-chambered exposure systems, and the techniques employed similar and standardized throughout the programs. The importance of this micrometeorology data is obvious in characterizing the growth and vigor of trees within the exposure systems, and this characterization of tree growth is imperative to document in future studies with these exposure systems. Standardizing the units for

Table VI-1. Components of pollutant exposure studies which should be standardized.

-
- ** POLLUTANT MONITORING
 - Equipment/Precision
 - Sampling frequency (dependent on deposition)
 - Handling missing data
 - ** MICROMETEOROLOGY DATA
 - Temperature
 - Solar radiation
 - Relative humidity
 - Wind speed
 - ** FORMATS FOR COMPUTER DATA BASES
 - ** UNITS
 - ** PERFORMANCE CHARACTERISTICS FOR DOCUMENTING SYSTEMS
 - ** EXPOSURE SYSTEMS
 - Dry deposition
 - Wet deposition
 - Wet/dry deposition
 - Particulate (need more information)
 - ** EXPOSURE REGIMES
-

various measurements and employing consistent formats for tolerance across all sites are obvious requirement for large programs where data from many sites will be integrated.

The desire to have researchers characterize their exposure systems and to maximize comparability of studies among research sites highlights the need to select performance characteristics that can document system performance. An extensive list of performance criteria are given in Table III-1 with three primary categories: (1) physical and chemical nature of the pollutant in the atmosphere of the exposure system; (2) physical and chemical features of the environment; and (3) biological features of the exposure environment. Documenting the biology of the trees might include whole plant carbon economy (i.e., carbon acquisition and allocation to various "sinks" and maintenance costs) as a measure of plant vigor in response to the exposure environment. This performance characteristic becomes critical because most exposure systems have been used with annual agricultural crops, not perennial, woody species, and not primarily in year-round studies of growth response. During the workshop, many considerations of exposure systems were tempered by the lack of plant (tree) performance data for the system. The atmospheric environment of the exposure systems is usually characterized, at least to some degree, by physical and chemical features; however, only minimal attention has been paid to plant performance, and there has been no substantial characterization of tree growth and tree culture needs.

Recommendations for exposure systems by categories of dry and wet deposition were made for the various types of systems; they were not directed toward a specific brand name or "official" equipment. Examples of the systems are given in references where appropriate, and potential users are charged to duplicate performance characteristics of the various type systems, not necessarily the specific equipment (brand-names, etc.) that comprise the system.

C. Dry Deposition

The participants recognized that the "ideal" system for multi-year growth studies with both seasonal and year-round exposure to different pollutant types would be structureless, i.e., not introducing artifacts of growth with hardware and shelter. There are, however, necessary tradeoffs. One is the need to exclude ambient pollutants (wet and dry deposition) and a chamber or enclosure is a means to accomplish this. Another is the need for a wide range of treatments in exposure-response studies and enclosures are a less costly means to accomplish this. The automated exclusion system, in use for some acid-rain deposition studies, represents an attempt to minimize the structural artifacts except during the ambient event (i.e., rain event). The cost of such systems, however, is not trivial, particularly when a range of treatments and replications are required.

Since there is no exposure system well-characterized for tree growth and pollutant delivery, participants recognized the value in using a variety of the available exposure systems. Thus avoiding the undesirable introduction of consistent bias into the programs yet the opportunity for cross-comparability between research sites with some systems is still possible. The workshop

recognized the utility of a variety of exposure systems for multi-year growth studies including both chambered and non-chambered types. Actually, since the exposure systems are all poorly characterized regarding use with trees, especially the larger sizes, a multi-system approach is appropriate, with characterization of the systems being conducted concurrently. If possible, developmental work on exposure systems should be funded. Recommendations for dry deposition exposure systems are given in Table VI-2. The chamber design recommended, based on years of experience in crop exposure studies and the attendant characterization of these chambers, was the open-top type. More important, it was recognized that the open-top chambers have the ability to exclude certain ambient pollutants, thus allowing a wide range of defined pollutant treatments with a moderate modification of the environment. Ambient pollutant are excluded with filters, although they are not available for all pollutant types. The charcoal filter used for ozone exclusion is rather non-specific, and the other ambient gases and particles excluded or not should be characterized. The availability of filtration systems for other gaseous pollutants and particulates in dry deposition studies is a recognized limitation of the open-top system, and filtration efficiencies and characteristics should be documented. The environmental modifications imposed by the chamber are relatively slight compared to other types of field chambers, and have been documented; consequently chamber effect on plant (annual crops) responses can be factored into the response evaluations. Documentation for tree species is not available. There are several acceptable open-top designs -- for example, the Heagle *et al.* (1973) chamber (Appendix C) has been used for a large variety of plant species with no consistent effect on annual crop growth during the growing season; the ITE chamber (Appendix C) has the most acceptable air flow characteristics of any current design, but is too small for many plant and tree species (D. Fowler, personal communication); the Kats *et al.* (1985) chamber (Appendix C) has proven useful for studies with small citrus trees. The much larger open-top chambers of the Boyce Thompson design (Laurence *et al.*, 1985) (Appendix C) may be useful for larger tree seedlings and saplings. Other chamber designs are appropriate for specific uses, such as those with umbrella tops for acidic rain and gaseous exposures (Hogsett *et al.*, 1985; Krause, personal communication) (Appendix C).

A limitation of the open-top chamber is the alteration of the microclimate, especially in the cooler months and during those particularly critical times of entering dormancy and bud break. The concern is one of uncertainty as to tree growth behavior with microclimate change. The chamber temperature increase of 1-3 C over ambient might alter metabolic activity in the trees and result in aberrant reaction to stress treatment. The possibility of winter desiccation exacerbated by continual operation of blowers for pollutant dispersal was a concern for proposed year-round exposure studies. These are all unknowns, and without data to predict the outcome it is necessary to encourage caution. Where possible, corroborative studies should be pursued to characterize these concerns. A corollary of the microclimate alteration is the uncertainty regarding the length of time trees should be held in the chambers or the duration of the exposure studies in open-top chambers. The altered microclimate might lead to cumulative effects on the tree growth/metabolism over time, and the nature of this effect is unknown. A third limitation of the open-top chambers is the size of the trees to be used. The maximum functional chamber size has not yet been determined, but the selection of the experimental

Table VI-2. Recommendations for dry deposition exposure systems.

Multi-year (2+ year) Growth/Physiological Studies:

Year-Round and Seasonal Exposures

OUTDOOR CHAMBERS (open-top design)

Strengths

- * Operational
- * Defined pollutant
- * Moderate environmental modification
- * Large range of treatments
- * Filtration and gradients defined

Limitations

- * Age/size of trees
- * Duration limit
- * Chamber microclimate (e.g., temperature difference)
- * Filtration not available for all pollutants

NON-CHAMBERED SYSTEMS (plume and exclusion)

Strengths

- * Minimum environmental modification
- * Supplementary
- * Large trees
- * Best for homogeneous canopy

Limitations

- * Needs development
- * Exclusion -- air flow across canopy atypical
- * Plume -- needs "clean" environment
- * Large space requirement for range of treatments

CUVETTES

Strengths

- * Supplementary to chamber or non-chamber studies

Limitations

- * Only a portion of tree exposed

Short-Term Growth/Physiological Studies:

OUTDOOR CHAMBERS (open-top types)

NON-CHAMBERED (plume and exclusion)

INDOOR CHAMBERS (all types)

CUVETTES (supplementary to outdoor systems)

Gas-Exchange/Pollutant Uptake:

INDOOR CHAMBERS (CSTR's)

CUVETTES

plants should ensure that they fit well within the chambers. Rapidly growing species will fully occupy present chambers within a few growing seasons. With more slowly growing species, there may be accumulated effects of the slight differences in microclimate that exist between chambers and the surrounding environment. There was concern and caution urged regarding size of trees to be used in chamber studies, however no guideline was established. Until documentation is available, caution is urged in using chambered systems for multi-year growth/physiological studies, and in particular year-round exposures. Additional concerns which might also present limitations in the use of open-top chambers for particular studies include: (1) suspected non-specific filtration since current systems (charcoal filters) may remove ambient gases and particles from the air in addition to the gas and pollutant under study; (2) the recognition that open-top chambers do not have the equivalent of ambient vertical gradients of pollutant concentration; and (3) the structural issue of chambers withstanding severe winter conditions for year-round exposures.

Non-chambered exposure systems were also recommended for multi-year studies. Although even less is known regarding the use of these systems with trees, the lack of structure-induced artifacts in the exposure studies is attractive and the minimum alteration of the environment is important for both year-round and seasonal exposures. Both types of non-chambered systems, the plume and air exclusion systems could be useful, depending on the specific research objective. The non-chambered systems can provide a supplementary approach to tree exposure studies using open-top chambers. The large circular or square plume systems which encircle the entire exposure plot are recommended for large-scale exposure of trees with no environmental modification due to the exposure system. The dispersion characteristics for the systems have been well designed based on computer modeling and field testing. The distance between emitters and receptor plants has been determined to provide uniformity in pollutant concentrations under ambient wind conditions. Development of computer-based pollutant dispensing, control and monitoring systems, and computer-based environmental monitoring permits continuous use of the systems and characterization of the pollutant exposures within experimental plots. Examples of this recommended plume exposure system are those of McLeod et al. (1985) and Greenwood et al. (1980). Aside from the lack of experience with the plume-type exposure systems for trees, a major limitation is the fact that only pollutant addition is possible. Consequently, a "clean" environment is required to achieve a full range of treatments or the effects of ambient pollutant conditions cannot be ascertained. Replication of treatments requires a large investment in space, not a trivial cost to consider in designing tree exposure studies. Homogeneous canopies are desired to avoid the air pollutant sampling discrepancies which could occur with heterogeneous canopies. The other non-chambered system recommended is the air exclusion design. With a high air flow and well-defined exclusion characteristics, it is recommended for both ambient air and pollutant addition type studies. Experience with these systems is limited to low-growing crops; performance with mid-size and larger trees is not available. These systems impose only minor environmental modification during the periods of exclusion with duct inflation. Examples of this type system include Kuja et al. (1985) and Thompson and Olszyk (1985). An important concern in use of the air exclusion system, like most chamber designs as well, is the atypical air flow across canopies.

The cuvette exposure systems for leaf, needle, or branch measurements were recommended for supplementary studies within both chambered and non-chambered systems. The cuvette systems facilitate more closely controlled pollutant exposures of leaves, needles, or branches in chambered and non-chambered systems, and would be especially useful with physiological studies of gas-exchange (photosynthesis, transpiration, pollutant flux rates) in larger trees, supplementing biomass studies. The pollutant exposures should be controlled to reflect experimental needs. It was suggested that additional work be conducted to evaluate the gas mixing and boundary layer characteristics of cuvettes, a more detailed analysis of the water vapor surrogate method of estimating SO_2 absorption by leaves, validation of this approach as a method for assessing the fluxes of other pollutants into leaves, and evaluation of the potential for using the water vapor surrogate approach (among others) to quantify the air pollutants adsorbed by plants in cuvettes, chambers, and chamberless plots. Selection of the appropriate cuvette type for the research objectives is important and is aided by reference to Table IV-5. Cuvette exposure systems are limited by having only a portion of the plant tissue being exposed to pollutant while the remainder of the plant is influenced by ambient conditions. This limitation is important in interpretation of these type studies.

The recommendations of the workshop participants for exposure systems to be used in short-term growth/physiology studies were the same as for multi-year growth studies, with the addition of controlled-environment or greenhouse exposure chambers. The modification of the environment inherent in indoor chambers is less of an issue for short-term studies of a few months, and is desirable for environmental or climatic interaction type studies.

Current technology will determine the capability to build the best possible indoor facilities that meet the basic requirements: (1) minimum fluctuation in temperature, humidity, and light conditions; (2) minimum gradients of pollutant concentration within chambers; (3) capability to create dynamic patterns of pollutants; (4) capability to determine pollutant flux rates; (5) capability to determine physiological parameters such as photosynthesis and transpiration rates; and (6) air movement for minimum leaf boundary layer resistance without wind injury.

Additional features recommended by Heagle and Philbeck (1978) (Appendix G) should be included: nonreactive surfaces; not a closed air flow system; negative pressure; easily portable (for some applications); and transparent covering for chambers within greenhouses or growth rooms).

All of these requirements can be met reasonably well by any of the four types of indoor chambers (Table IV-3). Chambers of the CSTR design (Heck *et al.*, 1978; Rogers *et al.*, 1977) are recommended for studies in which plant growth responses and gas exchanges are to be measured in well-mixed, spatially uniform gaseous environments. Growth room locations for CSTRs will be preferable when environment definition and repeatability are major factors in the research program; greenhouse locations will be adequate when the inevitable seasonal fluctuations in light and temperature environment are deemed acceptable in the specific experimental design. CSTR chambers should not be used when gradients of pollutant concentrations as environmental factors are desired. Non-CSTR chambers such as those described by Heck *et al.* (1968),

Jensen and Bender (1977), and Lockyer et al. (1976) appear to be as satisfactory as CSTR chambers for comparative studies of plant sensitivities to gaseous pollutants and are preferred over the CSTR chamber for studies involving gradients. Recently constructed self-contained units such as those of Aiga et al. (1984) and Payer et al. (1985) and chambers within rooms such as those of Payer et al. (1985) appear to establish the current state-of-the-art technology for indoor gaseous exposure chamber systems.

Gas-exchange/pollutant uptake studies should be conducted with indoor chambers of the CSTR-type (Appendix E) and cuvette exposure systems. Size considerations will limit the scope of such studies.

Screening-type studies can fall within both multi-year and short-term growth/physiological studies, and are essential to the Forest Response Program, both for screening sensitive species and/or clones, and screening effective pollutant type. The participants recognized that these screening studies could be conducted in both chambered (open-top) and non-chambered systems. Indoor chambers would also be useful, but only for short exposure periods of less than one year. Screening studies of growth response must measure current season growth and that following spring bud break, thus covering two growing seasons. Where physiological responses are used as a surrogate for growth there must be experimental evidence that establishes the relationship of the measured physiological process and growth (e.g., instantaneous measurements of net photosynthesis rate may not relate well to long-term growth).

D. Wet Deposition

The workshop participants identified certain elements in the monitoring of wet deposition studies and exposure systems that should be standardized; these are listed in Table VI-3. The frequency of these chemical analyses and measurements should be determined within the context of the quality assurance program development; more substantive characterization may be warranted with some experimental objectives.

Routine chemical characterization of the incident hydrometeor was strongly encouraged, and the characterization should include a complete cation and anion analysis. Similarly, regular characterization of the throughfall/stemflow was recommended, particularly if in the objective or design of the study a large percentage of the deposition is modified in passage through a canopy to the soil. Both the deposition rate and chemistry should reflect ambient conditions, depicting the full cation and anion features in addition to pH, nitrate, and sulfate concentrations. The dissimilar chemistry and deposition rate of cloud/mist versus rainfall should be recognized, in particular the enrichment of multiple cations, anions, and particles in cloud water relative to rainfall. When a soil component is included in the study, participants recommended that the porosity and solution infiltration rates be reasonable to allow for near normal chemical processing of the wet deposition within the soil and rhizosphere, and that the characteristics be documented.

Table VI-3. General recommendations for wet deposition exposure systems.

-
- oo Chemical analysis of the incident hydrometeor
 - oo Chemical analysis of the throughfall
 - oo Measurement of wet deposition rates
 - oo Determination of wet deposition chemistry
 - oo Inclusion of dynamic component in wet deposition exposure
 - oo Suitable physical and chemical properties of the soil
-

Recommendation of wet deposition exposure systems were a function of the experimental objective, tree or canopy height, site or location, and statistical design. Many of the same concerns raised with dry deposition were discussed, and the participants recognized that the "ideal" system would be one which introduces no artifacts from the physical structure of the system. Consequently, as with the dry deposition studies, no single system was recommended to accommodate all research plans. Experimental requirements in some instances dictate the need for exclusion of ambient dry and/or wet deposition and the ability to create a range of treatments for experimental design. In addition, available wet deposition exposure systems, like those used in dry deposition studies, have not been characterized regarding tree growth and vigor; consequently size/developmental stage of material, and duration of experimental time period is an unknown. Because of these and other uncertainties, it was recommended that a variety of systems be used until better characterization of the exposure systems with trees has been accomplished, perhaps in conjunction with planned experiments. As was suggested for dry deposition exposure systems, developmental work should be funded for various wet deposition exposure systems, especially cloud water deposition.

1. Rainfall Simulation

The workshop participants recommended the systems indicated in Table VI-4; each have merit for a particular application. The recommendations are made for either year-round or seasonal exposures with multi-year growth studies. The most flexible, in the sense of being able to address a variety of experimental objectives, is the automated exclusion system with modified scheduled addition of rain. The "ideal" system of no shelter (shelterless exclusion) is useful only in regions where the rain is not acidic, and where the volume of added rain would not exceed the normal range of rainfall for that location (i.e., the trees would not be overwatered). The automated exclusion system with the shelter coming into place only during ambient rain events is at this time the best compromise between chambered and non-chambered systems. There was the concern that since only rain is excluded in most of these type systems, there is the confounding influence of ambient dry deposition. It was recommended that continuous monitoring of ambient dry deposition be conducted for proper characterization of these exposure systems. The scheduled rather than automated addition of rain is recommended to allow for experimental flexibility. Scheduled additions should occur, however, during times when evaporation is low or there is not rapid evapotranspiration. The addition should simulate ambient rain events as recommended by the workshop (see exposure regimes below).

The shelterless and chambered system with permanent or manual exclusion of ambient wet deposition (Table VI-4) were preferred for certain experimental objectives and to address the limitations of the automated exclusion systems in multi-year growth studies. Rain addition was recommended as discussed above. The automated exclusion systems represent a significant financial investment in facility and equipment. They require some type of system to prevent interaction with dry deposition (e.g., chambered or duct-type air exclusion); they may not be capable of exclusion and addition with larger trees that have mature canopies. The shelterless systems have the advantage of minimum hardware, thus the least impact of structure on experimental design and would be the preferred

Table VI-4. Recommendations for rainfall simulation exposure systems.¹

Multi-Year Growth/Physiological Studies

o Automated exclusion/modified scheduled addition (ambient simulation)

Strengths

- * Reduces the structural artifacts of chamber on growth
- * Suitable for all experimental objectives

Limitations

- * Shelter must be in place during the "event"
- * No exclusion of ambient dry deposition
- * Significant financial investment
- * Questionable use with mature canopies
- * Questionable operation at high elevations, steep slopes, or high wind areas

o Shelterless exclusion/modified scheduled addition

Strengths

- * Minimum hardware, least impact of structure on growth
- * Suitable for mature canopies
- * Ideal for use in areas where rainfall not acidic

Limitations

- * Treatments have rainfall in excess of controls
- * No characterization of snow
- * Requires considerable water and mixing capabilities

o Shelter (permanent or manual) exclusion/modified scheduled addition

Strengths

- * Excludes some types of ambient dry deposition
- * Provides total exclusion of wet deposition
- * Useful in remote sites
- * Suitable for species and rain chemistry screening, climatic interaction

Limitations

- * Modification of growing environment with chamber

(continued)

Table VI-4 (continued)

Short-Term Growth/Physiology Studies

- o Same as for multi-year studies
- o Indoor chambers -- controlled environment or greenhouse

Strengths

- * Low cost
- * Variety of manipulation studies possible
- * Suitable for screening studies, physiology studies, climatic interaction

Limitation

- * Duplicate/terminal velocity of droplet.

¹ No priority implied in order of listing.

system for areas where rain chemistry is not seriously affected by anthropogenic inputs. The shelterless or non-chambered system is perhaps the most feasible for mature forest canopies and is suitable for species screening and/or study of rainfall chemistry. Caution was urged, however, regarding the addition of rain in excess of ambient to avoid complicating the interpretation of a stress response. With these systems, characterization of snow chemistry becomes a necessity. The shelterless plots, if large, require considerable water supplies and conditioning capabilities. The chambered systems (sheltered exclusion), in contrast, are most useful for excluding ambient dry deposition, and provide for total exclusion of any wet deposition. The permanent shelter can be used for species screening and testing various aspects of rainfall chemistry in areas with acidic rainfall. It is also valuable for testing interactive effects of wet deposition and natural climatic stress (e.g., drought, frost tolerance). In some locations, such as high elevation, slopes, or high wind areas, the chambered systems with permanent wet deposition exclusion might be the prudent choice. The primary limitation of the chambered system is the modification of the growing environment, and the degree to which this is crucial to interpretation of the experimental results has not yet been characterized. These same concerns relating to climatic modifications with chambered systems were recognized for dry deposition exposure systems. Supplementary studies are needed to answer the questions on how these systems affect tree growth and vigor. For year-round exposures, the issue of snow within the automated exclusion systems and the shelterless systems was discussed, but there was no resolution on how to handle this type of wet deposition.

Short-term growth/physiological studies should use the same systems as for multi-year studies; in addition, indoor chambers, either controlled-environment or greenhouse exposure chamber systems (Appendix E), are acceptable and in some instances desirable for screening studies, physiological studies, exposure-response, and climatic interaction studies. Indoor chambers are relatively low cost and there are many well-documented examples in the literature offering a wide breadth of possible manipulative studies involving climate, soil, species and rain chemistry. Caution is urged to control and characterize the degree and duration of foliage surface wetness by carefully maintaining appropriate temperature, humidity/evaporation conditions, and the realization that duplication of the terminal velocity of incident rain is probably not possible. Mimicking the physical characteristics of rain were discussed, but since there is no clear indication of the importance in wet deposition studies, recommendations for standardization were not made.

2. Cloud Water/Radiation Fog Simulation

Cloud water simulation studies introduce an even more complex element into exposure systems. The canopy impact of cloud water is an important component in this type of exposure. Thus, there is the need for delivery of the deposition with inertial velocity. The appropriate method was not resolved in the workshop; it was urged, however, that some effort toward development of exposure systems in this area be pursued, since the potential impact of cloud water chemistry, especially in high elevation forests, was recognized as crucial to an understanding of forest decline in these areas.

Recommendations were made regarding exposure systems that simulate radiation fog, where impact velocity is not a factor (Table VI-5). For multi-year studies, the open-top chambers were recommended primarily for the needed permanent exclusion, since no fog filtration is available and it is necessary to encapsulate experimental material for efficient delivery of a range of fog treatments. The chamber would be used with permanent or manually-movable tops in place during fog generation. The disadvantage is the modified environment and the extent of that modification has not been characterized for tree growth. There is also the need to control leaf surface wetness (evaporation). For short-term growth studies, controlled-environment exposure systems such as CSTR, as well as the greenhouse box-type chambers (Appendix E) were found acceptable. Wind tunnels (Appendix E) as exposure facilities for fog permit the characterizing of deposition, but have the disadvantage of limiting experiment duration since they are not constructed to optimize seedling growth. Also, the wind tunnel restrict plant material size to only seedlings. For cloud water simulation, the wind tunnel might be the optimum system for delivering inertial fog. Irregardless of exposure system, there is also the need to characterize the site of deposition within the canopy, especially with fog, in order to conduct appropriate monitoring of incident deposition.

E. Wet and Dry Combinations

The summary of workshop recommendations for combined wet and dry deposition exposure studies is presented in Table VI-6, and reflects the same strengths and limitations identified for the individual exposure systems. The recommendation of automated exclusion with scheduled additions for wet deposition, and a chamberless system for dry deposition offers the least structural influence on the exposure study. However, particular experimental objectives may be best served with one of the other two recommended system combinations; or the expense of the automated exclusion system may dictate an alternative approach. Lacking clear-cut characterization of the tree growth/vigor with these exposure systems, corroborative studies should be pursued with all systems. The open-top chambers are recommended for exclusion of ambient pollutant and because they offer the ability to deliver a wide range of experimental treatments. Scrubbing or excluding air pollutants limits the chamberless systems. The type of dry deposition pollutant under consideration in the interaction-type study would also affect the choice. For example, with ozone as the interacting pollutant, the open-top chambers with charcoal-filters are preferred; with SO₂ the non-chambered systems would be feasible, based on current knowledge of background SO₂ and frequency of occurrence. The short-term exposure studies using indoor chambers, both CSTR and greenhouse types, are recommended for screening studies and their ability to control evaporation more readily.

F. Aerosols

No recommendations were given for this type of exposure system since there has been little development based on experience of the participants and in literature reviews. Instead, the workshop participants recommended that monitoring for aerosols be included in the research plans. Monitoring is

Table VI-5. Recommendations for cloud water/fog simulation exposure systems.

Radiation Fog:

Multi-Year Growth/Physiology Studies

- o Open-top chambers (permanent or manual exclusion tops)

Strengths

- * Permanent exclusion (no fog filtration)
- * Encapsulates experimental material

Limitations

- * Modifies ambient environment
- * Control of evaporation

Short-Term Growth/Physiology Studies

- o Open-top chambers
- o Indoor chambers (CSTR, greenhouse, or wind tunnel)

Strengths

- * Control of evaporation
- * Suitable for species screening or deposition features
- * Climatic interaction

Limitations

- * Limited duration of experiment
- * Restricted to seedling size

Cloud Water Simulation:

Multi-Year

- o Nothing available

Short-term

- o Wind tunnel

Need developmental studies to characterize

Table VI-6. Recommended exposure system for combinations of wet and dry deposition.

Multi-Year Growth/Physiology Studies

- o Wet deposition -- automated or manual exclusion/scheduled addition
Dry deposition -- chamberless exposure system
- o Wet deposition -- automated or manual exclusion/scheduled addition
Dry deposition -- open-top chamber technology
- o Wet deposition -- permanent exclusion/scheduled addition
Dry deposition -- open-top chamber technology

Short-Term Growth/Physiology Studies

- o Same as multi-year plus indoor chambers
-

needed to examine the extent of occurrence and chemistry of aerosols. A similar recommendation was given for particulates in dry deposition since these two components of air quality may play important but as yet misunderstood roles in impact on forest growth, especially in high elevation forests and trees exposed to the vagaries of environment.

Summary

Workshop participants recognized the importance of heterogeneity in experimental approaches. In past, this recommendation responds to the lack of experience in documenting tree growth and physiology within the existing exposure systems. The workshop agreed that the "ideal" system would be one that introduces no artifacts on growth through structure of the system. In reality, however, there are particular research objectives that could best be addressed at this time with chambered systems which introduce the artifact of a structure in order to attain exclusion of ambient pollutants, create a range of pollutant treatments, or allow climatic interaction studies. In a number of instances, the issue of exclusion in experimental design, whether dry (gaseous) or wet, is best resolved with chambered systems. Documentation of tree growth in the open-top chamber and length of time for a growth study inside chambers is needed. Also, the experience factor with open-top chamber systems is important. The exclusion systems, either manual or automated, reduce the imposition of structure at least during an exposure event, and thus provide an advantage. However, the expense and the location requirements of the automated exclusion systems limit their use. Non-chambered systems (plume and exclusion) can also be quite costly, with extensive space requirements for treatments and replications.

The recommendation to employ both chamber and non-chambered systems for multi-year growth studies of wet and dry deposition also addressed the issue of introducing a consistent bias across all sites. Without knowledge of tree culture and exposure system types, the participants agreed that such a bias was not desirable. Cross-comparability would still be possible and supplementary studies should be designed within the National Forest Response Program four research cooperatives to address this issue by using similar systems and designs at different sites.

G. Exposure Regimes

Recommendations

The workshop participants addressed the question "Should exposure regimes be standardized?" They concluded were: (1) no, but there should be guidelines for developing exposure regimes; (2) exposure regimes should be region/site specific; and (3) exposure regimes should be selected to test experimental hypotheses.

The participants made eight specific recommendations regarding guidelines for regimes to be used in exposure studies within the Forest Response Program (Table VI-7). In addition, certain limitations were also recognized in current

Table VI-7. Recommendations for exposure regimes to be used in wet and dry deposition exposure studies.

-
1. The exposure regimes should reflect the episodic (seasonal, diurnal) nature of pollutant occurrence.
 2. The exposure regimes should be representative (frequency of occurrences) of the study area or the area where the study species is indigenous.
 3. The exposure regimes should consider the daily physiological function of the plant but the exposure should follow ambient air quality and not be arbitrarily terminated.
 4. The pollutant regimes should yield a range of pollutant(s) concentrations above and below the current ambient concentration(s).
 5. Exposure regimes should use realistic frequency and concentrations of pollutant co-occurrence.
 6. Data for the development of realistic exposure regimes for combinations of dry and wet deposition are limited.
 7. Data for the development of realistic exposure regimes for particles and aerosols are limited.
 8. Additional monitoring data are needed to characterize the concentration distributions for the dry deposition of particles and aerosols.
-

air quality monitoring for dry and wet deposition pollutants, and the need for additional monitoring data was noted as a priority within the exposure program (Table VI-7).

Participants recognized that the exposure regimes used in current gaseous pollutant studies could also be used to classify exposure regimes for other pollutant sources, i.e., wet deposition. The regimes are identified as either "artificial" or "episodic", with three variations on the episodic type. The definitions, recommended uses, and recognized limitations of each exposure regime type are summarized in Table VI-8 and discussed in Chapter 5. As with exposure systems, the choices of regime is dependent on the biological or research objective. Consequently, the participants made recommendations for the acceptable type of exposure regime based on the same generic biological objectives used in making the exposure system recommendations. The recommendations are summarized in Table VI-9 for the four different exposure regime types.

Recommendations for particular techniques to use in development and construction of exposure regimes were not offered. Some methods have been described in the literature for gaseous exposures, but this is a relatively new area of emphasis in experimental design and would benefit from new ideas. The methods currently used include both the characterization of ambient air quality by site and region, as well as the method for constructing experimental fumigation regimes (Male, 1982; Laurence and Kohut, 1984; Garsed and Rutter, 1984; Lefohn et al., 1986a,b).

Table IV-8. Types of exposure regimes currently used in fumigation studies with gaseous pollutants.

	Regimes		
	"Artificial"	Ambient	"Modified" Ambient Simulated Ambient
Definition:	Exposures were not designed to represent the temporal variation in concentration.	Exposures represent the temporal variation in concentrations.	Exposures represent the temporal variation in ambient concentrations.
Uses	- When insufficient monitoring data to characterize ambient air quality, or - Rapid-response monitoring equipment not available, or - Limited time resolution in the monitoring data. - Suitable for short-term studies and some screening studies.	- Assess the impact of current air quality at a site. - Suitable for long-term studies.	- Suitable for both long- and short-term studies in the field and lab. - Exposures can be replicated in time and space. - Can be used to develop exposure-response functions. - Can be used to create a range of concentrations. - Can be used to test hypothesis concerning the characteristics of exposure.
Limitations	- Not suitable for long-term studies. - Difficult to extrapolate the data to ambient conditions.	- Cannot be used to replicate the exposure in time or space. - Cannot be used to develop exposure-response functions.	- Not suitable for greenhouse and laboratory studies. - Exposures cannot be replicated in time or space.

Table VI-9. Summary of exposure regime recommendations for generic biological objectives.¹

Generic Objectives	Exposure Regimes			
	Artificial	Ambient	Modified	Simulated
Long-Term Growth/ Physiology				
Year-Round	?	Y	Y	Y
Seasonal	?	Y	Y	Y
Climatic	?	Y	Y	Y
Short-Term				
Growth	Y	Y	N	Y
Physiology	Y	Y	N	Y
Gas-Exchange	Y	?	N	Y

¹ Y = regime is appropriate.

N = regime not recommended.

? = regime is acceptable only if insufficient monitoring data to characterize air quality and/or rapid response monitoring equipment not available.

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